WSRC–MS–2000–00061

Proceedings of the Symposium
May 17, 2000

50 YEARS
of Excellence in Science and Engineering at the Savannah River Site

Savannah River Site
50th Anniversary

Prepared for the U.S. Department of Energy
Under Contract No. DE-ACO9-96SR18500
Westinghouse Savannah River Company
Savannah River Site, Aiken, SC 29808

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Preface and Acknowledgments

Throughout 2000, a remarkable variety of events are commemorating the 50th anniversary of the Savannah River Site. One of the major events is the symposium *50 Years of Excellence in Science and Engineering at the Savannah River Site,* sponsored by Citizens for Nuclear Technology Awareness and chapters of the American Nuclear Society and the Health Physics Society.

During most of its history, SRS was an important supplier of nuclear materials for America's nuclear arsenal. The Site also produced unusual radioisotopes in its reactors for research, industry, and the space program. At the end of the Cold War the reactors were shut down. Now the emphasis is on environmental remediation and the disposition of plutonium from decommissioned nuclear weapons.

In a technically challenging enterprise like SRS with its diverse nuclear facilities, a large community of skilled scientists and engineers is essential. This book, the symposium proceedings, highlights some of their outstanding achievements during the past half-century and some of the people associated with these accomplishments.

A principal purpose of the symposium is to bolster the institutional memory of SRS, not for fond reminiscence but as a foundation for vision and mission. Equally important, the proceedings can serve as a benchmark and reminder for the current and future technical staffs of SRS that they are members of a community with a long tradition of excellence in science and engineering. This should be a source of stimulation and encouragement.

The Symposium Committee invited current and former employees to nominate the innovations to be recognized. Several selection panels of experts in various technical fields reviewed 190 nominations and selected the achievements included in these proceedings. Neither the Symposium Committee nor the selection panels claim that these accomplishments are the best of the best. Instead, we believe they typify the outstanding quality of science and engineering at the Site during its first half-century.

Like most of SRS's achievements, many people worked together to make the symposium a success. First, we are indebted to the authors. About half of them have retired from SRS, some long ago. Our particular thanks to them for painstakingly reconstructing important technical programs in the earlier years of the Site.

Two of the authors, Dr. Myung Lee and Dr. D. Thomas Rankin, died several months before the symposium. The distinguished character of their work is shown in the symposium papers that describe their notable achievements.

The Symposium Committee acknowledges with gratitude the support of the Savannah River Operations Office of the Department of Energy and the Westinghouse Savannah River Company in the preparation of these proceedings. We also are grateful for the help from the 50th Anniversary Committee, chaired by John Granaghan, and the many organizations that provided financial support to that committee.

Special thanks to Fred Springer, Amy Phillips, and Cyndi Damiano for the editorial expertise needed to produce this volume.

Thanks to my fellow Symposium Committee members, Steve Epperson, Mel Buckner, Bruce Cadotte, Fred Springer, Todd Crawford, and Tom Parkinson for their time, effort, and good sense in making the symposium happen.

Finally, a blanket thank you to the many other people who lent a hand, especially those who nominated the achievements and the experts who served on the selection panels.

Bill Reinig

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The Genesis of the Savannah River Site Key Decisions, 1950

J. Walter Joseph and Cy J. Banick

Abstract

Key decisions that shaped the future of the Savannah River Site were made by members of the U.S. Atomic Energy Commission (AEC), the General Advisory Committee, and management of E. I. du Pont de Nemours and Company (Du Pont) during 1950. These decisions included:

- Defining the scope of the facility
- Selecting Du Pont as the prime contractor for design, construction, and operation
- Establishing appropriate AEC and Du Pont organizations to manage the project
- Choosing the location in South Carolina
- Defining the unique provisions of the Du Pont contract
- Agreeing on primary design criteria for the various site processes

The chronology of these decisions is summarized in Figure 1.

The decisions were made very rapidly by a small number of experienced and qualified people. The decisions were made boldly; many were based on data that were incomplete at the time. The effectiveness of the decisions was validated by the subsequent contribution of the Savannah River Site to ending the Cold War.

Background

The unexpected test of the first Soviet atomic bomb on August 27, 1949, shocked the U.S. nuclear establishment that believed it was several years ahead of the Soviets. “We will stop glorifying our past,” said Eugene Wigner, and the race was on.

Atomic Energy Commission Chairman David Lilienthal sent a letter to President Harry Truman in November proposing the development of a “super” bomb. The commission concluded that, with a minimum of three years of development, “there is a better than even chance it can be made to work.” On January 31, 1950, President Truman announced he had directed the AEC to continue work on all forms of nuclear activity, including the “hydrogen or fusion” bomb. The Commission and Department of Defense recommended a program for quantity production of materials for thermonuclear weapons, which was approved by President Truman on June 8.

As late as September 29, 1950, well after the program had been launched, minutes of the Atomic Energy Commission indicated that it was “impossible at this time to make a final determination of the feasibility of the tritium bomb.” This uncertainty was not erased until the Greenhouse test series in April and May of 1951 demonstrated thermonuclear principles. Thus, the decision to proceed with the project was a huge leap of faith and evidence of the perceived urgency.

Key Decisions

The Contractor

Du Pont was the leading candidate for the proposed project from the moment the concept was developed. Du Pont participation in the atomic energy program began in 1942 when Crawford Greenewalt was reassigned from his position as Technical Director of Grasselli Chemical Department to provide liaison with the atomic scientists working in the "Metallurgical Laboratory" at the University of Chicago, which was directed by Arthur Compton. The renowned physicist, Enrico Fermi, designed the world's first nuclear reactor at the Met Lab. On December 2, 1942, Greenewalt was present at the first self-sustaining chain reaction of the atomic "pile" under the stadium grandstand.

Greenewalt subsequently became Technical Director at Hanford when Du Pont was requested to design, build, and operate that plant. Greenewalt and Du Pont Chief Engineer Granville Read were cited by General Leslie Groves, who directed the Army's Manhattan Engineering District, as the two men "without whom we could not have completed Hanford." The ultimate accolade from the atomic scientists came in 1946 when Fermi asked Greenewalt to quit Du Pont and devote his life to pure research.

The company had continued to take an active interest in atomic energy after turning over operation of the Hanford Plant to General Electric in October 1946. Hood Worthington was one of the first members of the General Advisory Committee serving from 1946 to 1948 under Chairman J. Robert Oppenheimer. The Committee was formed to provide advice from experienced nuclear physicists to the AEC. Greenewalt and Donald Carpenter, vice president of Du Pont's Remington Arms subsidiary, were appointed to advisory boards in 1947 and 1948. In 1948, Du Pont was asked to study "all chemical activities bearing on the manufacture of plutonium" and make recommendations to the government. Du Pont performed this study at no cost to the government and continued to review Hanford operations and perform other studies requested by the AEC. Liaison offices were established at Hanford, Argonne National Laboratory, and Chalk River, Canada. These projects involved many Du Pont engineers and managers, including Monty Evans, Bill Mackey, Lombard Squires, Milton Wahl, Don Miller, and others who went on to lead the Du Pont Atomic Energy Division in the 1950s and beyond.

Preliminary negotiations with Du Pont were initiated as early as April 1950. Crawford Greenewalt had become president of Du Pont in 1948. Greenewalt was confident of Du Pont's ability to undertake the project because of the background of Hanford experience and demonstrated technical competence. The commission shared this confidence. The AEC considered briefly Union Carbide, Monsanto, Dow Chemical, and American Cyanamid for the project but concluded that Du Pont had unique qualifications and experience.

In a meeting on May 12, 1950, with Carleton Shugg, former AEC manager at Hanford, and Carroll L. Wilson, one of the original commissioners, Greenewalt insisted that a letter from President Truman endorsing the urgency of the project would be required to obtain Du Pont's participation and that Du Pont wanted full responsibility for the project, including design, construction, and operation. Greenewalt said the company would make no commitment until its engineers reviewed the AEC plans and evaluated the chances of completing the project on schedule.

In May 1950, Du Pont was asked informally to review technical aspects of the new project. This request was formalized June 12 when Acting AEC Chairman Sumner Pike requested Du Pont to review technical aspects of a new atomic energy production center for the purpose of considering a contract with the AEC for all phases of the work, including the site survey.

Curtis Nelson was appointed manager of the new AEC operations office for the tritium production project in June 1950. Like Greenewalt, Nelson was a veteran of the atomic energy business. He had broad construction experience and had been a colonel in the Manhattan Engineer District. After Hanford, he served as the AEC liaison officer at the Canadian Chalk River site where he became familiar with Canadian heavy-water reactor technology. (The 40 MW Canadian NRX reactor, fueled with natural uranium, moderated with heavy water, and cooled with light water, had been taken critical in 1947.) Nelson's deputy manager was Robert C. Blair.

After considerable discussion within AEC, Chairman Gordon Dean wrote President Truman on July 21 recommending that the president write to Greenewalt asking Du Pont to proceed with the project. President Truman wrote the letter on July 25.

Du Pont promptly formalized their commitment to the project by establishing the Atomic Energy Division (AED) within the Explosives Department. The AED management team was listed on the August 1, 1950, organization chart (see Table 1).

This organization was supplemented in a letter from Monty Evans to Curtis Nelson, AEC Operation Manager, on August 9. Additional assignments were V. R. Thayer, J. C. Woodhouse, D. F. Babcock, and C. W. J. Wende to the Research (Technical) Division and W. H. Holstein and J. B. Tinker to the Production (Manufacturing) Division.

AEC announced on August 2, 1950, that Du Pont had been selected as the contractor for design, construction, and operation of new production facilities to be built at a site yet to be determined. A letter contract was issued October 17 with an effective date of August 1. On the same day, Greenewalt wrote to President Truman to inform him of the contract and to assure him that Du Pont would "as always, put forth its best efforts."

The management teams brought to the new project by the Atomic Energy Commission and the Du Pont Company were experienced and well qualified to lead the new enterprise.

Site Scope

The initial budget proposal sent by President Truman to Congress in July 1950 was for two heavy-water reactors at the facility. In August, AEC told Du Pont that the Site should include five heavy-water reactors on normal (natural) uranium, a facility for Purex separation (Building 221), a fabrication facility for plutonium shapes (Building 235), a tritium separation plant (Building 232), and capability for irradiation of bismuth, if required.

The scope of work was modified in December 1950 to include addition of a second separation area with a future separation area (200-X) under consideration. In January 1951, the decision was made to build a heavy-water plant at Savannah River to supplement the Dana Plant. Du Pont recommended to AEC that the bulk of the electric power needed for SRP be generated on site in small, dispersed plants. In November

Table 1. AED Mangement Team, August 1, 1950

Assistant General Manager	— R. M. Evans
Administrative Assistant	— D. F. O'Connor
Atomic Energy Division Manager	— B. H. Mackey
Manufacturing Division, Director of Manufacture	— W. C. Kay
Control Division Manager	— F. M. Burns, Jr.
Technical Division Manager	— L. Squires
Assistant Manager	— J. E. Cole
Assistant Manager	— H. Worthington

1951, AEC eliminated funds for U-233 separation; plutonium production in all five reactors was recommended with excess reactivity applied to tritium production.

A power-producing heavy-water reactor was considered in initial planning. This concept was relegated to second priority at Argonne National Laboratory in February 1951 because of concerns that it might interfere with design of the production reactors. The sixth reactor was dropped from the budget in November 1952.

Facilities were added to and deleted from the scope of the site almost continuously as designs were firmed and requirements changed. This flexibility and adaptability became one of the principal attributes of the Site and allowed it to adjust to many changing missions through its first five decades of operation.

Site Selection

In June 1950, the Atomic Energy Commission asked Du Pont to locate a suitable tract of land for a plant to manufacture radioactive products. Originally, the study was to be limited to the "First Defense Zone," an area of the southeastern U.S. judged to be least susceptible to missile or sabotage attack from the Soviet Union. Basic site requirements were defined by Du Pont and agreed to by AEC:

- Manufacturing Area—Six reactor plant locations and one test location will be spaced approximately two miles apart and no closer than two miles to any other plant. Five separations plant locations will be approximately one mile apart.
- Site Area—The site area will include a 5.5-to-6-mile-wide zone outside the critical manufacturing area. All inhabitants or personnel not connected with the plant will be evacuated from the total site area.
- Supporting Population—The edge of the manufacturing area will be between 20.5 and 40 air miles to the edge of a center of population with at least 25,000 people.
- Isolation—Distances from the manufacturing area and maximum community populations shall be:

Distance (miles)	Maximum Population (persons)
5.5-10.5	500
10.5-15.5	5,000
15.5-20.5	10,000

- Water—Adequate water will provide cooling for six reactors without damage to other presently established users:

	Cooling Water (cubic feet/second)	
	Once-Through	Cooling Tower
Withdrawal for plant	600	180
Consumption	50	65

- Electric Power—Uninterrupted supply of 125,000 KW
- Railroad—Site must be accessible.
- Highways—Site must be accessible.
- Meteorology—No absolute limits but a favorable site would have prevailing wind velocities above 3 miles per hour directed away from centers of population closer than 20 miles. Climate should be as favorable as possible for plant construction and operation.
- Geology—The geological substructure and overburden should be stable with a low earthquake record and probability.
- Construction and Operating Costs—The Site should have characteristics that assure economy consistent with other requirements for satisfactory operation.

Eighty-four specific sites were identified in the First Defense Zone. Onsite inspections were performed by AEC, Corps of Engineers, and Du Pont representatives at 5 of the 17 most favorable locations. The study was subsequently extended to the Second Defense Zone, which included most of the northeastern, central, and southwestern U.S. to include sites with lower water temperatures and humidity. Six sites in this zone were visited, and the potential sites were reduced to four, two each from the two defense zones:

- Site Number 5—Aiken and Barnwell Counties in South Carolina, on the Savannah River, 20 air miles southeast of Augusta, Georgia, and 15 air miles south of Aiken, South Carolina
- Site Number 125—Fannin and Lamar Counties in Texas and Bryan and Choctaw Counties in Oklahoma on the Red River, 15 air miles east of Bonham, Texas, and 76 air miles northeast of Dallas, Texas
- Site Number 59—Crawford and Clark Counties in Illinois and Sullivan County in Indiana, on the Wabash River, 20 air miles southeast of Terre Haute, Indiana
- Site Number 205—Bayfield and Douglas Counties in Indiana, on the shore of Lake Superior, 26 air miles southeast of Duluth, Minnesota

Public Law Number 843 was passed in September 1950 to authorize AEC to acquire land for a plant to manufacture radioactive products. On November 10, Du Pont recommended selection of the Savannah River Site to the Site Review Committee, consisting of five members of leading engineering firms selected by AEC. Critical criteria were seclusion and an adjoining labor market; the dissolved mineral content of the Red River in Texas and construction difficulties in the northern part of the country also contributed. The committee, the Department of Defense, and the AEC concurred unanimously. The commission officially designated the Site as "The Savannah River Plant," and the Corps of Engineers was authorized to acquire approximately 240,000 acres. The public announcement of the Savannah River Plant was made November 28.

The commission expressed concern over the original plan, which included the sites of the towns of Dunbarton, Ellenton, Jackson, and Snelling within the plant's boundaries. By year's end, the project boundary line was changed to exclude Snelling and Jackson. The layout provided space for five reactors with two additional sites available. The first property was transferred December 29, 1950, and all SRP land, including 6,000 acres around Lower Three Runs Creek, was acquired by June 30, 1952.

The total impact of the land acquisition included:

- 1,500 people
- 1,706 tracts of land
- 200,742 acres
- 165 cemeteries containing 6,000 graves; 124 cemeteries with 4,980 graves were removed and reburied.

The Contract

Du Pont had negotiated a unique relationship with the Army Manhattan Engineering District for their work at Hanford. Notable features of the contract were:

- Du Pont received a fee of only one dollar for what became a half-billion dollar project.
- Du Pont continued to apply corporate pay scales rather than government pay scales to employees who were transferred or hired for the project. This permitted Du Pont to assign its best people without sacrifice because corporate pay scales were 150%-250% higher than government or university pay scales for equivalent work.
- The government reimbursed Du Pont for all costs and losses incurred as a result of the work.
- The government took possession of all products. This was important to Du Pont because most of the products were dangerously radioactive.
- Du Pont retained the option of leaving the enterprise nine months after the war ended. This option was invoked in October 1946, 11 months after the Japanese surrender was signed.

Crawford Greenewalt took a firm position on the proposed contract for the new production facility, insisting that it be modeled on the Hanford relationship. There was some reluctance within AEC to follow the Hanford model, but Du Pont stood firm. Greenewalt explained the Du Pont position to the Joint Committee on Atomic Energy on August 4, 1950. He pointed out that Du Pont did not seek the assignment and would undertake it only because of a clear need on behalf of national security, as demonstrated by the July 25 letter from President Truman. He described the decision to do the work for no fee as resulting from two considerations: (1) the experience that Du Pont brought to the job had been gained at government expense, and (2) Du Pont felt that " . . .we simply cannot be in a position of making money out of an engine of war that is as horrible as this one is likely to be." The Du Pont president told senators and congressmen that Du Pont expected to be reimbursed by the government for all reasonable costs. He said general overhead costs would be held as low as possible, as was the case in normal commercial practice.

Greenewalt devoted most of his presentation to a discussion of the importance of paying employees in accordance with normal Du Pont compensation practices. He said Du Pont intended to staff the plant with "our very best people" and that they would be compensated on the same terms as they would have been had they remained in commercial activities. Greenewalt ended his discussion by stating that Du Pont had concluded that the elements of the project appeared to be perfectly feasible.

Despite some continued resistance within the commission, Gordon Dean wrote President Truman on September 27 requesting the president to "authorize AEC to proceed with a contract with Du Pont that would be similar to the Hanford job." Letter Contract AT(07-2) was issued to Du Pont on October 17 with an effective date of August 1. The contract contained all of Greenewalt's key requirements. The contract allowed Du Pont the freedom to recruit competitively and carry out the project in accordance with established corporate practices. In today's legalistic environment, it is hard to imagine that a major corporation would perform work for the government for six months with only a "handshake agreement" rather than a binding legal contract.

Process Designs

Reactor

In February 1950, the Joint Committee on Atomic Energy (JCAE) discussed four alternatives for producing tritium:

- Load the H Reactor at Hanford with enriched uranium.
- Build six materials testing reactors (MTRs). The MTR at Idaho Falls was a 40 MW reactor with enriched fuel, cooled and moderated by light water, was about to be built, and would be taken critical in 1952.
- Use a large linear accelerator being studied currently at Berkeley Radiation Laboratory.
- Continue design work on a large heavy-water-moderated reactor similar to the NRX, which had been in operation at Chalk River, Canada, since 1947.

The JCAE concluded that the last alternative seemed to be efficient and realizable and an ad hoc AEC committee, chaired by George Weil, recommended that the heavy-water reactors be built to produce materials for thermonuclear weapons. On July 20, Du Pont concurred in the selection.

Eugene Wigner, the Nobel-prize-winning theoretical physicist had proposed light-water cooling for the Hanford reactors and advocated heavy-water moderation in the early 1940s. The basic concept for a reactor cooled and moderated with heavy water had been developed by the prolific Walter Zinn at Argonne National Laboratory (ANL). Zinn worked closely with the Canadians, who had heavy-water operating experience with their NRX plant. Argonne expanded its heavy-water reactor program rapidly during 1950 to develop experimental data on reactor physics and engineering. Much of the work focused on the metallurgy of reactor materials.

The AEC initially established an objective of approximately 1800 MW of total reactor capacity as necessary to produce the quantities of tritium thought to be required. They decided subsequently that the Savannah River reactors should be scaled at 300 MW. Thus, six reactors were proposed originally, and the original site layout included six reactor plants. The August 1950 scope of work called for five reactors.

Du Pont proceeded rapidly with detailed design of the reactors. The design team placed "a large premium on flexibility in the ultimate design." This flexibility was required because of AEC uncertainty as to the relative quantities of plutonium and tritium that were required. In January 1951, Du Pont reported that the reactor design could incorporate flexibility "without loss" for either plutonium or tritium production.

Du Pont arranged with ANL to place young engineers for training and work in physics, chemistry, engineering, and metallurgy at Argonne. Milton H. Wahl, who later was appointed director of the Savannah River Laboratory, led the Du Pont Argonne group. By August 1951, 66 Du Pont employees were working at ANL.

The emphasis on reactor flexibility produced versatile machines capable of operating at powers almost an order of magnitude higher than the design basis and producing isotopes not yet discovered in 1950.

Separations

Substantial work on processes for separating desired isotopes from irradiated reactor components had been conducted at Hanford, Oak Ridge National Laboratory (ORNL), and Knolls Atomic Power Laboratory (KAPL). The original separations plants built at Hanford used a bismuth-phosphate co-precipitation process that was capable of recovering plutonium but not the large quantities of uranium that went into the waste tanks with highly radioactive wastes. The inefficiency of this process was well understood at the time, but the Army pressed for the simple process because of the wartime urgency. Recovery of the uranium became essential later because of dwindling supplies.

After the war, Hanford worked on the Redox solvent extraction process. In 1948, AEC requested Du Pont to collect information related to recycling uranium and handling fission products and wastes. This endeavor was led by Monty Evans, who later became the first assistant manager of Du Pont's Explosives Department with responsibility for the Atomic Energy Division. Du Pont recommended development of Redox, but work at Hanford proceeded slowly, and the Hanford Redox plant did not start up until August 1951.

Meanwhile, ORNL and KAPL had developed an alternative solvent extraction process known as "Purex" that used a less flammable solvent and produced a substantially smaller volume of liquid wastes. Although Purex was not as well developed as Redox; it had been tested thoroughly on a laboratory scale by the time Du Pont assumed the contract for the new facility. Du Pont immediately dismissed several other separations processes as being unable to assure "a reasonable chance of operating successfully." The selection of the Purex process over the Redox process was recommended in a letter from F. S. Chambers to Lombard Squires, dated September 27, 1950. A large Du Pont group was established at ORNL under Luther Peery and Bob Martens; a smaller group was installed at KAPL.

The Purex process worked well at Savannah River, and Purex variations are the international standard for production and power reactor fuels.

Heavy Water

Heavy-water-cooled and -moderated reactors required a large amount of heavy water, typically 250 tons per reactor. The total world supply of heavy water in 1950 was less than 50 tons. Production of a large volume of heavy water was a major challenge in the early stage of the project and was expected to be on the critical path. Three processes were available for heavy-water production:

- Distillation and catalytic exchange of light water
- Low-temperature distillation of liquid hydrogen
- Gaseous hydrogen sulfide/liquid water dual-temperature exchange

The distillation process had been used by Du Pont during the Manhattan Project to produce about 30 tons of heavy water. However, this process was prohibitively expensive for the large volumes needed for the new reactors.

After the war, AEC asked Hydrocarbon Research, Inc., to design a plant based on the hydrogen distillation process. Despite concerns about the hazards of handling hydrogen gas and operating difficulties with the low-temperature process, the commission approved construction of a pilot plant on March 1, 1950.

Early work on the dual-temperature process had been done under Harold Urey at Columbia University. The process used hydrogen sulfide gas, which was very toxic, in pairs of alternately placed hot and cold mixing towers. Scaling up the process from the laboratory bench to production presented engineering difficulties, but the AEC contracted with the Girdler Corporation to use some of the existing facilities at the Wabash River Ordnance Works near Dana, Indiana, and to build a pilot plant. Inclusion of heavy-water production in the pending Du Pont assignment was considered by the AEC in May 1950.

Du Pont initially had concerns about heavy-water production based on their experience with the distillation process. Further studies convinced them that the dual-temperature process was the best option because of its cost benefits. Greenewalt recommended dual temperature to the commission on July 20, 1950. Du Pont was authorized to deal directly with Girdler and proceed with construction of a heavy-water plant on September 29. They recommended building six production units at the Site in addition to the Girdler pilot plant.

In recognition of the importance of the Wabash operation, the commission renamed the heavy water portion as the Dana Plant and established an area office, reporting to Curtis Nelson, in October 1950. The Dana pilot plant completed its first test run October 26. Many potential operating problems were solved there, and the nucleus of the Savannah River heavy-water operating staff was trained there. In January 1951, Du Pont was authorized to build Savannah River heavy-water production lines similar to those at Dana.

Despite initial concerns, 250 tons of heavy water had been produced before the first reactor (R) was completed.

Appreciation

The authors searched primary and secondary references for information on the formative decisions related to the Savannah River Site. The references were selected from materials collected by Mary Beth Reed, Steve Gaither, and Mark Swanson of New South Associates as part of the Site History Project under U.S. Department of Energy Cooperative Agreement DE-FC09-97SR18903. The draft of this paper was reviewed and suggestions made by several former Site employees.

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Biographies

J. Walter Joseph

Mr. Joseph graduated from North Carolina State in 1950 with a BE in Mechanical Engineering. After serving in the Army during the Korean conflict, he joined the staff of the Department of Engineering Research at Penn State and received his MS in Mechanical Engineering in 1954.

He worked for Du Pont and Westinghouse at the Savannah River Site from 1954 to 1993. He started in the Pile Engineering Division of the Savannah River Laboratory, where he conducted research in reactor heat transfer and hydraulics, stress analysis, and effects of irradiation on structural materials. In 1965, he transferred to the Savannah River Plant where he held a variety of technical and management positions in several departments, including Reactor Technology, Equipment Engineering, Traffic and Transportation, L Startup Project Team, and Site Quality.

He currently is founder and principal consultant of Quality Partners, providing training and consulting on Total Quality processes.

C. J. Banick

Mr. Banick earned his MS degree in Physical Chemistry from Georgia Tech. He currently is serving as WSRC's liaison to the SRS History Project. Previous experience at the Savannah River Plant and Laboratory included (1) assistance to plant startup and trouble shooting chemical problems associated with production reactors, (2) analytical chemistry support of operating and R&D efforts (principally by non-routine analytical methods development) in fields of reactor productivity, raw materials, fuel reprocessing, isotopic power sources, environmental monitoring, and characterization of special reactor irradiations, (3) writing, editing, producing, and distributing technical documents, (4) as classification officer, responsibility for approval processing of technical documents prior to public release, (5) as patent liaison, assisting DOE patent counsel in identification and prosecution of patentable inventions, (6) initial involvement in technology transfer activities, and (7) coordination of information exchange between WSRC and the Defense Nuclear Facilities Safety Board.

Figure 1. The Genesis of the Savannah River Site

	Oct.	
1949	Nov.	< AEC letter to President Truman proposes development of "super bomb".
	Dec.	
	Jan.	
	Feb.	< President Truman directs AEC to work on "hydrogen or fusion" bomb.
	Mar.	
		< AEC recommends heavy-water reactors for new plant.
	Apr.	
		< AEC initiates preliminary negotiations with Du Pont.
	May	
1950	Jun.	
		< Du Pont asked to locate a suitable site.
	Jul.	
		< Du Pont recommends dual-temperature heavy water production.
	Aug.	< Scope of project defined to include five reactors. < AEC announces selection of Du Pont as contractor.
	Sep.	
		< Du Pont recommends Purex separations process.
	Oct.	
		< AEC letter contract issued to Du Pont.
	Nov.	
	Dec.	< Public announcement of Savannah River Site is made.
	Jan.	
	Feb.	< Official start of construction.
1951	Mar.	

Heavy Water for the Savannah River Site

J. W. (Bill) Morris
William P. Bebbington, Robert G. Garvin, Mal C. Schroder,
and W. C. Scotten

Abstract

In late 1950, the supply of heavy water was recognized as a critical factor for the early startup of proposed nuclear reactors at the Savannah River Site (SRS). A new production process was demonstrated using hydrogen sulfide in a dual-temperature cycle with water to extract and concentrate heavy water from natural water. Two large plants were built and operated beginning in 1951. One was located at the Dana Plant in Indiana, and the other was at the Savannah River Site (then the Savannah River Plant). Formidable problems were resolved in control of the new process, in dealing with the corrosive hydrogen sulfide-water system, and in handling large amounts of the hazardous gas. Tonnages of heavy water produced were unprecedented at the time and were available early enough that supply was not a limiting factor in reactor startups. Both plants operated with excellent safety records and with high on-stream efficiencies. With adequate supplies of heavy water on hand and with no significant new requirements in the United States, the Dana Plant was closed in 1957. A major part of the SRP heavy water production plant was closed later in 1957-1958, and the remainder shut down in 1982.

This will be an historical saga, not a technical paper. Heavy water was a critical item for Site reactor startups, and that need was met. Between 1951 and 1982 over 7,000 tons was produced by Du Pont.

Water is a chemical compound consisting of two atoms of hydrogen and one of oxygen, designated in chemical shorthand as H_2O. Hydrogen is the smallest and simplest of all the chemical elements and was assigned an atomic mass of one. In fact, however, hydrogen in the earth's environment contains three isotopes, forms of the element that behave almost the same chemically but have different masses. Nearly all natural hydrogen does have a unit mass of one. But about 1 part in 7000, or about 140 parts per 1,000,000, has a mass of 2 and is called deuterium, symbol D. The third isotope of hydrogen has a unit mass of 3 and is called tritium. Tritium is radioactive, and its concentration is negligible in natural sources.

The term "heavy water" refers to deuterium oxide, D_2O. In contrast, natural water is called "light water" because the hydrogen in natural water is more than 99.98% mass 1. The heavy-water concentrations of natural waters do vary a bit from the normal range of 0.0148 ± 0.0002 %, but no natural enrichment is known that would be economically significant.

The production program for heavy water for the Savannah River Plant (SRP) began before the Site was chosen. Du Pont had pertinent experience with heavy water, having produced 32 tons of it for the Manhattan District in facilities at 3 ordnance plants built and operated by the Company during World War II. These large facilities used vacuum distillation of water followed by electrolysis for final concentration. When the Atomic Energy Commission (AEC) in the late 1940s undertook a large program to produce both tritium and larger amounts of plutonium, heavy water was the preferred moderator for the new nuclear reactors. Three production processes were seriously considered for the most difficult step; increasing the initial 1 part in 7000 by a factor of several hundred; vacuum distillation of water, distillation of liquid hydrogen, and exchange of liquid water with hydrogen sulfide (H_2S) gas in a dual-temperature cycle. Vacuum distillation of water was known to work but at great cost.

Hydrogen distillation offered a large separation factor but handling large quantities of liquid hydrogen had not been demonstrated. The large quantities of hydrogen needed were not readily available. Hydrogen sulfide dual-temperature exchange was feasible based on laboratory data, but the process had not been demonstrated. A critical problem in process control had been recognized but not solved, and the gas was both corrosive and very toxic. The potential advantages of the hydrogen sulfide process were so significant, however, that in 1949 the AEC asked the Girdler Corporation to design, build, and operate a pilot plant of significant scale and to design a large production facility. This program was given high priority because availability of unprecedented quantities of heavy water would be critical to the schedule for the proposed nuclear reactors. The location chosen for the pilot plant and the potential production plant for the "GS" (for Girdler sulfide) process was the site of the Wabash River Ordnance Works, which included the largest of the WW II heavy water plants. It offered some equipment that might be reused, and a large steam generating plant that could be returned to service.

When Du Pont accepted its role as prime contractor for what became the Savannah River Plant project, responsibility for heavy-water supply was included. An early review confirmed the advantages of the H_2S process if major uncertainties of toxicity, corrosion, and process control could be overcome. At this point Du Pont joined Girdler with primary responsibility for oversight of design and development activities, including operation of the pilot plant to demonstrate operability and process control. Du Pont's experience with hazardous materials gave optimism that dedicated safety procedures and equipment could handle gas toxicity, and an extensive corrosion research program was initiated within the Du Pont Engineering Research Laboratory to confirm and extend preliminary conclusions that conventional materials of construction could be used. Du Pont engineers worked with Girdler on process design and materials of construction. Girdler engineering personnel were very capable and cooperative, and this collaboration worked well. Construction of the pilot plant was nearing completion and about a dozen Du Pont people were transferred there to oversee operations. We were there on duty in late November 1950, when the Savannah River Site was announced. That heavy-water facility soon became known as the Dana Plant, named for the nearby village of Dana, Indiana.

The GS process is based on the fact that in the gas-liquid H_2S-H_2O system, deuterium is distributed better to the liquid phase at low temperature than at high. Thus, if a stream of water flows first down a cold gas-liquid multicontact tower countercurrent to a flow of hydrogen sulfide gas, then down a similar hot tower countercurrent to that same gas, deuterium will be absorbed from the gas in the cold tower, stripped from the liquid in the hot tower, and thus concentrated at the bottom of the cold tower and the top of the hot. Portions of those concentrated streams can be withdrawn from that two-column "stage", either as product or for further concentration. The process control problem is that the liquid/gas (L/G) ratio of these countercurrent flows must be controlled more closely than conventional instrumentation allows. Deviation of the L/G from optimum, either up or down, greatly decreases productivity. The solution to the problem was devised by the late Dale F. Babcock, senior member of Du Pont's pilot-plant task force. He pointed out that the mid-column concentrations of deuterium in the hot and cold towers would be nearly equal at optimum L/G, and that the ratio of the two concentrations would be extraordinarily sensitive to variations in the L/G. Use of this principle in the pilot plan demonstrated that it solved the problem, and the ratio was later used routinely in the production facilities. Analysis was made by mass spectrometer to determine mass 18:19 (H_2O:HDO) ratio, and at higher concentrations for mass 19:20 (HDO:D_2O).

The pilot plant had been designed and built as a four-stage unit. The major uncertainties were process control, operability, and tray efficiency under process conditions. Resolving these uncertainties required only the first stage, consisting of 2 mild steel towers, each 3 feet in diameter by 110 feet tall and with 70 bubble cap trays. Auxiliary equipment included a rotary sliding vane compressor for gas circulation and centrifugal pumps for liquids, with spares in both services. A particular problem was that hydrogen sulfide and water form a solid hydrate at temperatures below 29.5ºC (80ºF) at 325 psig. The hydrate problem required that all sample lines and other small lines be heated to at least 30ºC (86ºF). Steam tracing was used for small lines, and generally electrical heating for instrument enclosures. This hydrate problem was a real aggravation for attempted pilot-plant operations during November 1950, when ambient temperatures dropped to −20ºF (-29ºC).

Shortly after pilot-plant startup, hydrogen sulfide stress corrosion cracking became painfully evident when the internal roller bearings of both the gas blower and its spare shattered into sharp, hard steel fragments. Despite this and numerous other problems, successful operation of the pilot plant was achieved beginning at about 2 p.m. on October 26, 1950, and continued for almost 300 hours before shutdown was necessitated by other mechanical difficulties. During this operation, the concept of the ratio of mid-column concentrations to indicate the critical liquid/gas flow ratio was well demonstrated. The data showed that a flow ratio near optimum was maintained from hour 100 to 140, and that directions and approximate magnitudes of needed corrections in flow ratios were clearly indicated. The data also permitted determination of average bubble cap tray efficiencies of at least 45% for the two-column system. Taken as a whole, the pilot-plant operations and data were judged adequate to justify the choice of the GS process for the production plant. The operational difficulties, however, together with the risk of loss of valuable product by high pressure leaks, led to the decision to limit that process to about 15-20% heavy water. This approximately thousandfold factor over the initial concentration of 0.014% constitutes the bulk of the total separative work and consequent cost for both facilities and energy. Vacuum distillation (DW process) would be used to bring the concentration up to about 90%, followed by batch electrolysis (E process) to achieve final, reactor-grade purity of 99.75% minimum. These two processes represented extensive industrial-scale experience, simple and straightforward process control, and little possibility for product losses. At Dana, parts of the wartime plant were used for the DW process.

The Girdler Corporation continued with engineering design and began construction of the Dana Plant production facilities. Particular attention was given to the intricacies of process control, corrosion/materials of construction, and the hazards of hydrogen sulfide. Shortly thereafter, decision was reached that an additional heavy-water plant also would be required to meet the schedules for the new nuclear reactors. That plant would be built at the Savannah River Site, along with a power plant to provide both electric power and process heat.

The R&D program on materials of construction was expanded and carried out throughout design and construction, the results being applied concurrently at both plants. Subsequent findings in the operating plants extended those experimental data. The following principles were used in construction and maintenance as early as possible:

1. Carbon steel was used for most process vessels and for heat exchanger shells and piping. Steel plate to be used for process vessels was carefully examined by ultrasonic inspection to reject any that contained fissures, voids, or laminar inclusions. Most bubble caps and trays were constructed of Type 410 stainless steel (SS). At SRP, however, towers in two of the three “buildings” were clad and caps and trays were constructed with Type 304 SS.
2. Hardness of all bolts was limited to 27 on the

Rockwell "C" scale, and imposed stresses were limited to 40,000 psi measured by extensometer as bolts were tightened.

3. Process units were to be given thorough annual inspections and hydrostatic testing.
4. Lower-than-normal industrial velocities were used in steel piping, and liquid entrainment was minimized in gas lines.
5. Stainless steel was used where high velocities were necessary (e.g., orifice plates).
6. Minimum thickness holes (small holes drilled partway through) were used to give early warning of wall thinning where erosion or entrainment was likely.
7. Metal parts in which stress or hardness was necessary, such as Bourdon tubes, springs, and instrument bellows, were isolated from hydrogen sulfide.

While the foregoing measures did not eliminate corrosion in the process plants, they did make the consequences tolerable.

Beyond the foregoing measures that were taken to contain the hydrogen sulfide, a broad program was established to deal with its inherent hazards. The material is extremely toxic, more so than hydrogen cyanide. Each of the two GS plants would contain about 800 tons of the gas under pressures up to 250-300 psig. The physiological effects of H_2S are insidious in that the gas has a foul odor of rotten eggs at initial exposure to low concentrations, but continuing exposure to higher concentrations anesthetizes the olfactory system and masks the odor. Exposure to significant quantities of the gas can quickly lead to unconsciousness, but recovery is rapid and complete if fresh air is provided quickly. To deal with these hazards, a comprehensive safety program was formulated, including the following:

1. Extensive monitoring systems were established to detect hydrogen sulfide in the air.
2. Masks with breathing air reservoirs ("Air Packs") were provided in process areas and absorbent canister masks in more distant areas. All personnel in plant areas were trained in artificial resuscitation.
3. A 400-foot-high flare stack was provided for each GS plant to vent and burn gas that had to be released. Quick-acting isolation and dump valves were provided.
4. A "buddy" system was established for the GS and gas generation plants. All personnel working in the units were extensively trained and worked in pairs. Each individual in the pair carried an air pack, and they stayed far enough apart that if one were overcome, the other could rescue him.

These measures, and constant vigilance, permitted the entire production program of heavy water to be carried out with no serious injuries from exposure to hydrogen sulfide. A few people were overcome, but all responded either to fresh air or to artificial resuscitation if necessary, and none suffered any lasting ill effects.

Engineering design for the Dana production facility proceeded with the concept developed by Girdler under its initial contract with AEC. Extraction and initial concentration of heavy water from the feed water from the Wabash River involved six GS units, each consisting of five stages of cold-hot tower systems. The first stage of each unit consisted of four cold-hot tower pairs in parallel; the second stage, one cold-hot tower pair of the same size; and subsequent three stages, towers of the same height but progressively decreasing diameters. Each tower had 70 trays and was about 120 feet tall. The first- and second-stage cold and hot towers were 11 and 12 feet in diameter, the third 6 and 6.5, the fourth 3.5 and 4, and the fifth 2.5 each. Concern for the operability and process control of the large, complex tower systems of the Dana design led to the choice of simpler independent units for the plant at the Site. It had 24 identical units, each with only 2 stages, but the second stages each had over twice as many separative trays as those in the Dana design. The second-stage SRP cold and hot towers were each built as two physical units in series and were 6.5 feet in diameter, roughly the same as stage 3 at Dana. The first stage SRP towers were the same diameters as those at Dana. The diameters of the larger towers were limited to sizes that could be transported by rail from fabrication plants scattered about the country. Fabrication of the 96 towers for Dana and 144 for Savannah River taxed the capability of the vendors of such equipment in this country during construction of the two plants. Space does not permit inclusion of photographs or flow diagrams, but each of the two large plants was quite impressive in appearance.

Each of the 24 GS units of the SRP design contained only 4 interrelated process flows, while each unit of the Dana design contained 6 times as many. Flow control problems at Dana were compounded by the much more complex interstage relationships and by the parallel first-stage arrangements. Because of these complexities, all gas process blowers and liquid circulatory pumps at Dana were equipped with spares to minimize upsets, while spare blowers and pumps were not justifiable in the simpler SRP design.

The first attempted startup of a Dana Plant GS unit occurred during the winter of 1951-52. Hydrogen sulfide gas was introduced to displace water in the first stage towers. After the system had been brought up to process conditions and flows, pressure drops in the towers were lower than expected, and anticipated buildup of heavy water was not achieved. When the unit was shut down and the towers were opened for inspection, many of the uppermost trays in the towers were found to have collapsed. Evidently, the temperature had fallen so low that solid hydrogen sulfide hydrate formed, overloading and collapsing the trays. Also, many of the slotted bubble caps on the trays were broken. The caps had been made by cold pressing, and had not been annealed. Much work and several months of delay were incurred while the column internals were removed and repaired, bubble caps and other manufactured parts were replaced by properly stress-relieved parts, startup procedures were extensively revised, and piping changes were made to accommodate the revised procedures. Such lessons learned at Dana were very painful, but the resulting findings were beneficial to both heavy-water plants. The first successful startup of a Dana GS unit was in early August 1952, and of one at Savannah River was in October of that year. All parts of both plants were in full operation by May 1953.

The Dana Plant provided the nucleus of the operating and technical staffs at Savannah River, and the two plants collaborated fully in all matters. In fact, the plants engaged in friendly but vigorous competition, especially in safety records and productivity. Each plant had been designed for a conservative production capability of 240 tons of heavy water per year. Each achieved that rate within about a year after full startup, and exceeded it by more than a factor of two within about two more years. Both plants operated consistently well after a variety of startup problems. Changes were made when indicated. In one instance, after a number of years of operation, the screwed joint of a 16-inch flange failed at SRP and within 20 minutes 46 tons of H_2S burned before it could be dumped to the flare tower. The heat carried the gases up above the surrounding towers where winds dissipated the fumes. H_2S was barely detectable at ground level, and no injuries resulted. Investigation found that poorly machined threads and warping of the flange by normal bolt loads caused the failure. Thereafter, all 16-inch and 12-inch screwed flanges were replaced by welded flanges.

Process control using ratios of mid-column concentrations worked well at both plants. The sensitivity of that ratio was so high that a change of flow rate as small as 0.5% could be detected and correction could be made. Over the course of time and with instrumentation steadily improving with experience, sampling frequencies were considerably reduced.

The simpler design of the GS process at SRP led to lower down time and higher operating efficiency there. The SRP GS units were out of productive operation only about 2% of the time, most of which was for the required annual overhaul, inspection, and hydrostatic test. The more complex Dana units, in contrast, required about 17 days, or about 4-1/2% of annual operating time. Also, at Dana these procedures were prohibitively difficult during the cold winters, whereas at SRP such work was possible year-round. Thus, the productivity at SRP was greater than at Dana.

The production capabilities of the two heavy-water plants had been planned to meet the needs of the new reactors at the Savannah River Site without the heavy-water supply becoming a limiting factor. That goal was accomplished, and with a large reserve on hand, the Dana Plant was closed early in 1957. Although the facility had operated satisfactorily, major replacement of corroded Type 410 stainless steel trays would have been needed soon, and operating costs were higher than those at Savannah River. In the course of time, the Dana facilities were dismantled and sold.

At Savannah River, one of the three GS buildings, the one with unclad towers and Type 410 SS trays, was shut down on October 4, 1957. Corrosion was much more severe than in the other two buildings with clad towers and Type 304 SS trays. Soon afterward, calculations showed and a plant test demonstrated that with only two GS buildings operating, the E Plant could be shut down, and final product concentration was achieved in the DW Plant without significant loss in production. Operation without the E Plant produced considerable cost savings.

In October 1958, one of the remaining two GS buildings was shut down as inventories mounted. Also, to reconcentrate heavy water diluted during reactor operations, and to avoid tritium contamination of virgin heavy-water product, 3 of the 10 DW towers were isolated for such "rework" in 1957 and a fourth was added in 1960. Ultimately, the need for new heavy water decreased, and inventories rose to the point that this remaining production unit was shut down on January 8, 1982. At that time only three reactors were in service, their losses of heavy water were small, a large inventory was on hand, and continuing production was not justified. The "SRS News" of December 1995 carried a story and photograph of the dismantlement and removal of the flare stack, the GS process facility, the hydrogen sulfide generation plant, and interconnecting piping. Thus were removed the last visible indications of the production program for heavy water for the Savannah River Site.

Several corollary activities of the production program that warrant mention were carried out by or for the AEC and its successor agencies. Considerable amounts of heavy water were sold to scientific organizations for research, and by 1964, almost a thousand tons had been sold and exported to other countries. Several countries seriously considered heavy-water-moderated reactors for electric power generation, and permission was given for some of them to send visitors to the SRP production facility to obtain first-hand information. Canada in particular, having built a unique facility that produced a significant tonnage of heavy water for the Manhattan District during World War II, established a major program for producing, using, and exporting heavy-water-moderated power reactors. From 1965 to 1973, the Canadians built three different large production plants based on the GS process. The first suffered major deficiencies in design, and Du Pont engineers were called upon to help in devising remedies. The plant was ultimately rebuilt completely. Later plants were successfully built and operated after Canadian personnel were trained at SRP. Du Pont engineers were assigned to assist in startup and initial operations of these Canadian plants. Over time, these plants produced considerably more heavy water than had the U.S. plants. But as in the U.S., eventually decreased demands and increasing inventories led to shutdowns. As of this writing, the last of the Canadian plants are being dismantled.

Last but not least, major credit for the successful production of heavy water at both Dana and Savannah River goes to the people who oversaw operations and the maintenance work, and in general made the production wheels go around. They seldom had their names attached to technical papers, but without them the plants could not have operated safely or successfully. Their work in dealing with hazardous gas under high pressure, their dependence on the buddy system, and the stressful work of annual overhauls melded them into cohesive, effective units.

In summary, heavy water was produced for the Savannah River Site in two major production facilities designed, built, and operated under Du Pont supervision. Over 6000 tons were produced at Savannah River, and an estimated 1,200 to 1,500 tons at the Dana Plant. The plants operated safely despite handling unprecedented quantities of extremely toxic gas, and their product was available in time for the demanding schedule for reactor startups.

Biographies

J. W. Morris

J. W. (Bill) Morris earned B.S., M.S., and Ph.D. degrees in chemical engineering at the University of Texas at Austin. He then joined Du Pont in Wilmington, Delaware, but shortly was assigned to Du Pont's work for the Manhattan Project. After training at Oak Ridge, he went to Hanford, heading radiation protection for the final purification, concentration, and shipment of the product, plutonium. In late 1945, he returned to Du Pont's Grasselli Chemicals Department, where he served in Wilmington and in Cleveland, Ohio. In 1950, he joined Du Pont's return to nuclear matters, first in heavy water production at the Dana Plant in Indiana, where he became Technical Superintendent. In 1953, he came to the Savannah River Laboratory in South Carolina. There he was successively Director for separations R&D; for reactor engineering and materials R&D; and for University Relations. After a final assignment in environmental engineering, he retired at the end of 1983.

W. P. Bebbington

William P. Bebbington earned a Ph.D. degree in chemical engineering at Cornell University in 1940. In addition to his heavy-water experience during World War II, he held Du Pont assignments in high-pressure synthesis of ammonia, methanol, and polyethylene. As Du Pont began work on the Savannah River Plant in 1950, he headed the plant's Heavy Water Technology Section and later held management assignments in research and development. During Mr. Bebbington's last 12 years at SRP, he was General Superintendent of the Works Technical Department with responsibility for all aspects of the nuclear fuel cycle and also for radiation protection and environmental monitoring and protection. He has published technical papers on the production of heavy water, nuclear criticality safety, and environmental effects. He is the author of a *Scientific American* article on reprocessing of reactor fuels and a booklet on radioactive wastes that has been widely distributed by the American Institute of Chemical Engineers. He is a fellow of the American Institute of Chemical Engineers and in 1979 received the AICHE's Robert E. Wilson Award in Nuclear Chemical Engineering.

R. G. Garvin

Robert G. Garvin earned a B.A. degree and a B.S. in chemical engineering degree at Rice University and a M.S. in chemical engineering from North Carolina State University. He joined Du Pont at the Savannah River Plant in 1957 and served in a number of technical and management positions, including 10 years with heavy water operations, until his retirement in 1991. In 1969, he and the late Charles Gresham served as technical and production advisors respectively at the heavy-water plant in Port Hawkesbury, Nova Scotia, during startup and initial operation. From 1970 to 1973, he served as SRP liaison at the Bruce Heavy Water Plant in Douglas Point, Ontario, during the final stages of construction and startup.

M. C. Schroder

M. C. (Mal) Schroder earned a B.S. degree in chemical engineering from Louisiana State University in 1950 and joined Du Pont in Wilmington in 1951. Shortly thereafter he was called to serve in the Korean conflict. He returned to the Du Pont Construction Division at Pensacola, Florida, in 1953. The next year he transferred to the Savannah River Plant where he was assigned to the Separations Technology Group. In 1960, he was assigned as a supervisor in the Heavy Water Technology Group, and he later became superintendent of the Heavy Water Production Group. When heavy water production was curtailed he was transferred to other positions in Plant Maintenance, Defense Waste Planning, and design liaison and plant startup preparations for the Defense Waste Processing Facility. He retired at the end of 1986, but was called back as a consultant for the design of a new production reactor in 1991-92.

W. C. Scotten

W. C. Scotten earned a B.S. degree in 1943, and a Master of Science degree in 1947 in chemical engineering from the University of Missouri-Columbia. After joining the Pure Oil Co. in 1943, Mr. Scotten joined the Naval Reserve from 1944 to 1946. He subsequently worked for Du Pont at the following locations: Du Pont Grasselli, Experimental Station, Wilmington, 1947-51; Du Pont AED Works Technical, Dana, 1951-52; Du Pont AED, Works Technical, SRP, 1952-1982.

Development of Coextruded Fuel and Target Tubes for the Savannah River Plant Reactors

Philip H. Permar

Abstract

The coextrusion process devised by Nuclear Metals, Inc., was adapted to manufacture the 14-foot-long tubular fuel elements for the Savannah River Plant (SRP) reactors. Initially, smooth, aluminum-clad, uranium-aluminum alloy fuel tubes were fabricated. Subsequently, for high-power operation, many different ribbed fuel and target tubes were produced for use at SRP. A computer program was finally created for the design of extrusion dies used for the large-scale production of ribbed tubes.

Background

The designs of the first fuel and target assemblies at the Savannah River Plant (SRP) were based on Du Pont's prior experience at the Hanford Plant with short, aluminum-clad cylinders of natural uranium metal called "slugs". In the initial operation at SRP, four columns of similar short slugs were assembled in 14-foot-long vertical "Quatrefoil" tubes that controlled the flow of heavy-water coolant past the slugs. Hundreds of these fuel assemblies were arranged in a vertical lattice in the reactor. It was recognized that assemblies of Quatrefoil tubes would soon be replaced by fuel assemblies designed for operation at higher power.

Early in 1951, the Du Pont Atomic Energy Division selected Nuclear Metals, Inc. (NMI) as a metallurgical consultant because of NMI's experience in manufacturing fuel elements for a variety of U.S. reactors. NMI also pioneered "coextrusion" for producing reactor fuel elements at SRP. The process simultaneously formed the tubular fuel core and bonded the aluminum cladding to the inner and outer surfaces of the tube, hence the term "coextrusion".

Coextrusion Process Development

Development of Smooth Fuel Tubes

After several years of consultation and countless experiments at the Savannah River Laboratory and Plant (SRL/SRP), the first experimental coextruded smooth fuel tubes were produced in Building 320-M in September 1956. One year later, Building 321-M was completed for the fabrication of highly enriched tubular fuel, and these facilities were used for the remainder of the coextrusion program.

The coextrusion process at SRP employed a conventional 3000-ton extrusion press widely used in the aluminum industry. The extrusion press included an electrically heated container that held the hollow extrusion billet and a hydraulic ram that pushed the billet through a specially shaped die. The Moczik Tool and Die Company of Detroit, Michigan, cooperated with NMI and SRL to design and manufacture the extrusion tooling.

The keys to the development of a satisfactory smooth coextruded fuel tube were the end shape of the uranium-aluminum alloy core and the selection of the aluminum alloy used for the end plugs inside the composite billet (Bebbington 1990.) With properly designed billet components, the coextrusion process produced a smooth extruded tube containing acceptably shaped core ends.

The composite billet also included an evacuation tube to remove air from the assembly before extrusion. The extrusion lubricant, first lead-oil and later tin-oil, was specially selected to promote streamlined flow of the billet components. The outgassed billet was lubricated, heated, and extruded to produce a smooth aluminum-clad coextruded tube. After the tube had cooled, it was cleaned, straightened, cut to length, bond tested, and attached to the end-fittings to form the finished fuel element assembly.

Development of Ribbed Fuel and Target Tubes

Smooth coextruded fuel tubes proved to be excellent sources of neutrons in the SRP reactors. But when the demand for tritium continued to increase, the reactors were required to run at substantially higher power, far beyond the capacity of a single smooth coextruded fuel tube, to remove the heat from each vertical assembly.

The physicists quickly generated many designs for assemblies containing multiple fuel and target tubes that would run at higher power to produce a variety of radionuclides. As many as three fuel tubes and one target tube were included in a single assembly that fit into the original 4-inch-diameter position in the reactor lattice. These new designs required that the concentric fuel and target tubes be spaced by longitudinal ribs that ran the full length of the tubes.

For several years, ribbed tube extrusions were produced using smooth, unribbed billets. In these extrusions, the core was slightly thickened at the base of the ribs, but the local increase in heat generation was tolerable.

By early 1967, again with the help of consultants from NMI and Moczik Tool and Die Company, an improved manufacturing process was developed by SRL/SRP to produce the ideal ribbed tubes. Integral longitudinal ribs were created by using a ribbed extrusion billet in conjunction with specially shaped grooves in the extrusion die that formed the ribs on the tubes. Considerable development was required to establish the relationships among dimensions and shape of the ribs on the billet, the billet container, and the extrusion die as well as other extrusion parameters. For example, the lubrication of the billet and die with a tin-oil mixture was important in producing smooth surfaces, not only on the tube, but also on the sides and tips of the ribs.

In the beginning, the design of ribbed extrusion dies was more an art than a science. By 1967, extensive work had shown empirically that the material to form the ribs should be taken from the outer cladding, but the design and manufacture of a satisfactory die was a problem.

The major achievement at SRL was optimizing the design of the coextrusion dies for a variety of ribbed tube designs. These dies produced tubes with no core thickening at the base of the ribs. Eventually, a computer program was developed to produce the die designs.

Further technical details of the coextrusion process are discussed in Chapter 8 of the *History of Du Pont at the Savannah River Plant* (Bebbington 1990). This chapter contains an excellent description of the process, and also notes "The precise uniformity of thickness of core and cladding achieved in these fabrications is a truly remarkable feat of metallurgy."

Contributors

Assistance in the preparation of this paper was provided by the following individuals, most of whom worked in the Nuclear Materials Division at SRL:

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Frederick C. Rhode

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Biography

After graduating from Cornell University in 1943 with a Bachelor's degree in Chemical Engineering and a minor in metallurgy, Philip H. Permar was hired by Du Pont Engineering Department and worked at the Experimental Station in Wilmington, Delaware, on metallurgical problems in various Du Pont plants. He transferred in 1950 to the Du Pont Atomic Energy Division where he worked on design, construction, and process development of 300 Area at SRP. In 1952, he transferred to Aiken, South Carolina, as manager of the Nuclear Materials Division at SRL to work on the development of fuel and target elements to meet the changing demands for production of nuclear materials at SRP. In 1969, he transferred to the market development program for the neutron-emitter californium-252, a product of SRP, and promoted uses for californium-252 in the U.S. and Europe. Mr. Permar retired in 1982 after 39 years of service with Du Pont.

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High-Performance Uranium-Metal Fuels for Savannah River Reactors

William R. McDonell, George R. Caskey, and Carl L. Angerman

Abstract

The development of high-performance uranium-metal fuel elements for Savannah River reactors required a major metallurgical effort to achieve satisfactory irradiation behavior. Fuel element designs were progressively modified from the original small-diameter slug through a hollow slug of similar dimensions to the final large-diameter tubes that provided the increased heat transfer surface needed for high reactor powers. Concurrent adaption of cladding techniques led to the development of hot-press bonding and later to hot-die-size bonding of the aluminum can onto the nickel-plated uranium core to replace the original aluminum-silicon-dip bonding process.

Concurrent modifications in processing the uranium cores were necessary for satisfactory reactor performances of the fuel elements. An intermediate rate cooling process, specifically oil quenching, for beta host treatment of the alpha phase cores was developed to prevent introducing (a) crystallographic texture due to fast cooling from the beta phase, which resulted in anisotropic shape changes during irradiation, and (b) a large grain size from slow cooling, which produced surface roughening. Low solubility alloying elements, such as silicon and aluminum, were added to the uranium to control the gross cavitational swelling encountered during irradiation at elevated temperatures. The resulting uranium fuel elements proved capable of sustaining the reactor exposures required for many years production of nuclear materials at the Savannah River Plant.

Introduction

Uranium-metal fuel elements used in the heavy-water-moderated-and-cooled Savannah River Plant (SRP) reactors evolved over the initial 20 years of reactor operation through a series of changes in design, composition, and fabrication processes. These changes were dictated by production requirements, cost reduction, and fuel-element performance. This paper outlines the principal technological features of this evolution, emphasizing the metallurgical development required to produce a high-performance fuel element.

Fuel Design and Fabrication

The fuel elements employed originally in the SRP reactors on startup in the early 1950s were 1-inch-diameter x 8-inch-long solid unalloyed uranium slugs machined from rolled rods and clad in aluminum cans for corrosion protection from the coolant water. To improve heat-transfer capability, this design was changed in the late 1950s to a hollow slug of similar external dimensions. The slug was clad with aluminum on both outer and inner surfaces. Further improvement in heat transfer was realized in the early 1960s by introducing nested assemblies of large-diameter (2 to 4 inches) tubular elements with extruded uranium cores and aluminum cladding. Typical fuel configurations are shown in Figure 1.

The fuel elements were fabricated from uranium cores produced at the Fernald Plant, National Lead Company of Ohio, by shaping cast billets at high-alpha-phase temperatures and heat treating at beta-phase temperatures.[1] The heat-treated cores were clad in aluminum cans, which were metallurgically bonded to the core to ensure good heat transfer during operation and to limit core corrosion if the cladding were to be penetrated, as at a closure defect.

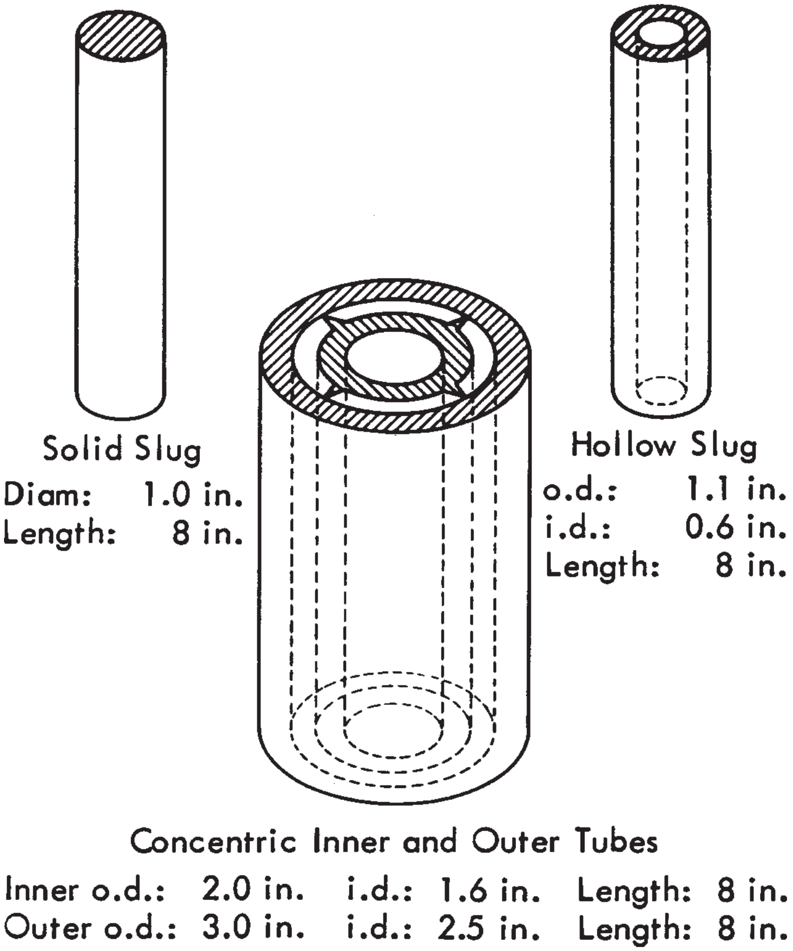

Figure 1. Typical configurations for SRP uranium fuel elements

Three techniques, Al-Si dip canning, hot pressing, or hot-die sizing, were employed to bond the aluminum cans to the uranium cores.

The solid slugs and some of the hollow slugs were canned by the Al-Si dip process, in which the heated slug was inserted into the aluminum can under the surface of a molten Al-Si bath, and an aluminum endcap was added. This assembly was then quenched in water. Bonding resulted from the thin Al-Si braze layer. Final closure was by tungsten-inert-gas (TIG) welding the can to the endcap.

Before canning, the uranium slugs were given a beta-phase heat treatment in a molten bronze bath, followed by quenching in a molten tin bath. This heat treatment was originally included as a preliminary step in the canning operation, but in later Al-Si dip-canning processes the treatment was performed during slug fabrication.

Most of the hollow slugs and all of the large-diameter tubular fuel elements were canned by hot-pressing or hot-die-sizing techniques. Both processes required a previously beta-treated core, electroplated with a 0.0001-to-0.0005-inch nickel layer, to serve as a bond between the uranium and the aluminum.

In the hot-pressing process, the electroplated uranium core was placed in a preformed aluminum can, an aluminum endplug was inserted, and the can was pressed mechanically at elevated temperatures to achieve bonding.

The hot-pressing operation produced bonding by interdiffusion at the uranium-nickel and the nickel-aluminum interfaces to form well-defined intermetallic compounds. Final closure was by welding.

The hot-die-sizing process was developed specifically for canning large-diameter tubular elements. In this process, the nickel-plated uranium core was loaded into the aluminum can, an aluminum endcap was inserted, and the preheated assembly was forced through a die to size the can onto the core. The die-sizing operation reduced substantially the thickness of the can wall and formed a diffusion bond between the uranium and aluminum components and the intermediate nickel layer. The ends of the element were bonded by a hot-pressing operation. Final closure was by welding.

Bond strengths of the hot-pressed and hot-die-sized elements were about twice that of the Al-Si-bonded elements. Bond strengths were used as indicators of good heat transfer for the elements. Special advantages of the hot-die-sizing process were the high rate of throughput of canned elements, small capital investment, and low cost of operation.

Fuel Element Structure and Irradiation Behavior

Tailoring the metallurgical structure and properties of the uranium core was necessary to achieve satisfactory fuel performance under the increasingly severe irradiation conditions imposed by the optimized fuel designs. In SRP reactors, the fuel elements were stacked in vertical columns within aluminum-alloy process tubes, through which the relatively low-temperature coolant water (D_2O) flowed. Coolant-channel spacing was maintained by longitudinal ribs on the process tube and fuel-cladding surfaces. The coolant-channel spacing limited the amount of distortion of a fuel element that could be tolerated during irradiation without disruption of reactor operation.

The most important sources of dimensional change during irradiation of a uranium fuel element were anisotropic growth and related surface roughening and swelling. Anisotropic growth—a change in shape with no large change in volume—and surface roughening were direct manifestations of the basic dimensional instability of the orthorhombic uranium crystal under irradiation. Polycrystalline specimens with crystallographic texture would undergo dimensional changes during irradiation analogous to those of the alpha-uranium single crystal (i.e., lengthening in predominantly *b*-axis directions, shortening in *a*-axis directions, with essentially no change in *c*-axis directions). Random textures were required to avoid anisotropic growth. Moreover, a specimen with large grain (or grain-domain) size would suffer a surface roughening during irradiation because of the same instability in the individual grains; therefore, a small final grain size was required.

Swelling, a volume increase of the fuel element during irradiation, occurred as the result of agglomeration of either irradiation-generated lattice vacancies into cavities and voids at intermediate temperatures or fission gases into bubbles at higher temperatures. The swelling at intermediate temperatures could be greatly enhanced by stress from the interaction of individual grains undergoing anisotropic growth during irradiation. Agglomeration of fission gases constituted a less severe problem in SRP reactors because operating temperatures were below the gas-swelling range.

Anisotropic Growth Control

Satisfactory resistance of the SRP fuel elements to anisotropic growth was attained by adopting heat-treatment processes that provided a randomly oriented, small-grained uranium structure. Experience prior to startup of the SRP reactors had led to the development of beta-phase heat treatments to avoid, on the one hand, the anisotropic growth of uranium caused by texture produced by forming in the alpha phase and, on the other hand, the excessive surface roughening caused by the large grain size of cast or gamma-formed metal. The beta treatment, incorporated initially as a preliminary bronze-tin-dip in the Al-Si canning process, yielded a structure with acceptable dimensional stability for early SRP operation.

The introduction of hot-press bonding a few years later, however, required a prior beta treatment, which was initially carried out in molten salt with water quenching. Application of this process to hollow slugs led to the incidence of anisotropic growth caused by texture produced in the fuel core during fast cooling from beta-phase temperatures. The hollow slugs, heat treated using a water-quench process and canned by either Al-Si-dip or hot-pressing procedures, increased in both inside and outside diameters, as well as length, and decreased in wall thickness, by a much greater extent than slugs heat treated by the previous bronze-tin-quench procedure.

The as-worked texture of the metal formed in the alpha phase was usually eliminated during beta treatment. Conditions of the beta treatment, particularly the means of cooling from beta-phase temperatures, had to be controlled, however, so as not to reintroduce the texture (McDonell and Sturken 1966). Fast cooling, as produced by water quenching, was found to produce a pronounced texture, with the *a* axis of the uranium crystal oriented predominantly in the direction of heat flow during cooling, and the *b* axis oriented predominantly in other directions. Thus, a hollow slug quenched from beta temperatures exhibited predominantly *a*-axis textures in radial (wall thickness) directions and *b*-axis textures in longitudinal and circumferential directions. Such a texture accounted for the anisotropic growth of the water-quenched hollow slugs during irradiation. Slow cooling, as in air, produced a large grain size in the metal, causing surface roughening during irradiation. The slow cooling could also produce a texture in the fuel, though resultant anisotropic growth was obscured by the surface roughening.

Intermediate-cooling-rate treatments were found necessary to obtain an adequately small grain size without excessive texture. Irradiation tests, conducted in conjunction with texture and metallographic studies, correlated the behavior of hollow slugs and tubular fuel elements with several candidate, intermediate-cooling-rate treatments. Of these, the oil-quench treatment was generally the best. It yielded a moderately small-grained microstructure with relatively little texture and resulted in near-optimum stability during irradiation.

Swelling Control

Fuel-core splitting, which caused slug-type elements failure, led to considerable cooperative efforts among Hanford, Savannah River Plant, Mallinckrodt, and National Lead before SRP startup to reduce levels of impurities that might embrittle the metal. This effort was successful, and this good-quality unalloyed uranium was in use as fuel in SRP reactors when preliminary irradiation tests of the large-diameter fuel tubes revealed a new and unexpected form of dimensional instability (Angerman and Caskey 1964). This instability was caused by formation of large (100 mm) internal cavities in the fuel at intermediate temperatures (see Figure 2). Tubular fuel with large cross sections of metal at intermediate temperatures swelled 10% or more. This swelling produced principally wall-thickness increases that caused detrimental coolant-flow changes and increased the potential for cladding failure. Below the intermediate temperature range, only minor volume increases (<1%, attributed to formation of solid fission products) were observed; above this temperature range, large brittle cracks, but no cavities, were seen.

The intermediate temperature swelling, termed “cavitational swelling,” differed considerably from the hitherto recognized swelling of fuel materials caused by the formation of fission-gas bubbles at high temperatures. Fission-gas bubbles in SRP fuel were shown to contribute only a small volume increase (<1%). Formation of the large cavities was therefore attributed to internal interactions between individual grains undergoing anisotropic growth in the metal.

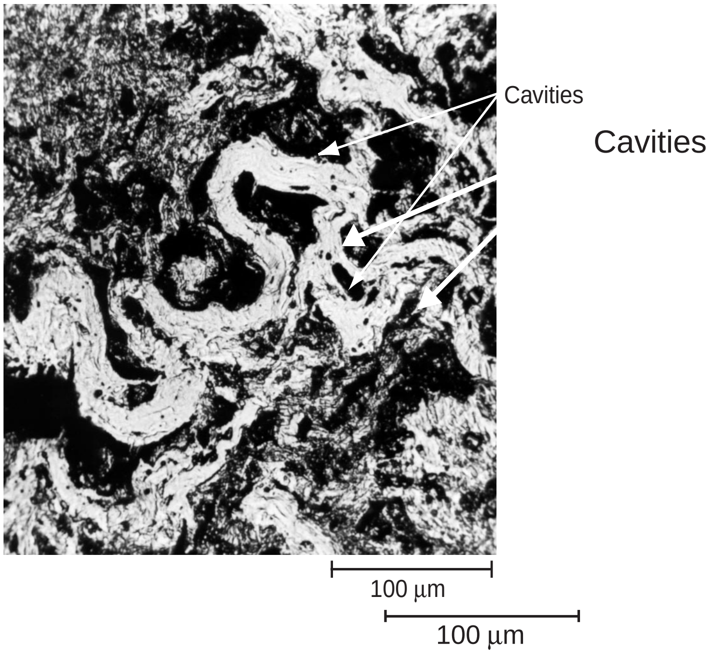

Figure 2. Large cavities formed in swollen regions of uranium fuel elements

Cavitational swelling produced significant volume changes only after critical exposure thresholds were exceeded (see Figure 3). The exposure threshold for unalloyed uranium decreased with increasing temperature in the intermediate temperature range, but at higher temperatures, apparently increased again, in accordance with the decreased susceptibility of the metal to anisotropic growth.

Observations that swelling behavior varied markedly among fuel elements derived from different billet castings suggested that relatively minor variations in impurity levels affected cavity formation. Impurity silicon in the range 10 to 65 ppm appeared especially significant. Irradiation tests of such variables demonstrated that minor additions of iron and silicon (100 to 150 ppm) markedly improved fuel behavior by extending exposure thresholds for cavitational swelling (see Figure 3). These results agreed with Harwell reports of the good behavior of uranium fuel containing small iron and aluminum additions. SRP fuel specifications were accordingly modified to provide for additions of 125 to 200 ppm of iron and 75 to 150 ppm of silicon, and these values were later increased to 125 to 225 ppm of iron and 125 to 225 ppm of silicon.

Additional irradiation tests over the period, undertaken in conjunction with microstructural and mechanical testing studies, served to define the swelling behavior and its relation to the structure and properties of a large number of Fe-, Si-, Al-, Cr-, and Mo-containing alloys, for exposures up to 5000 MWd/t at temperatures up to 500°C (see Figure 4) (McDonell, et al. 1969). These results were later extended to greater than 10,000 MWd/t at temperatures up to 400°C. Testing specimens at high exposures was generally undertaken in NaK-containing stainless-steel capsules, where detrimental dimensional changes could be accommodated without interference with reactor operation. These tests were followed by irradiation of satisfactorily performing compositions as full-size tubular fuel to verify expected performance characteristics.

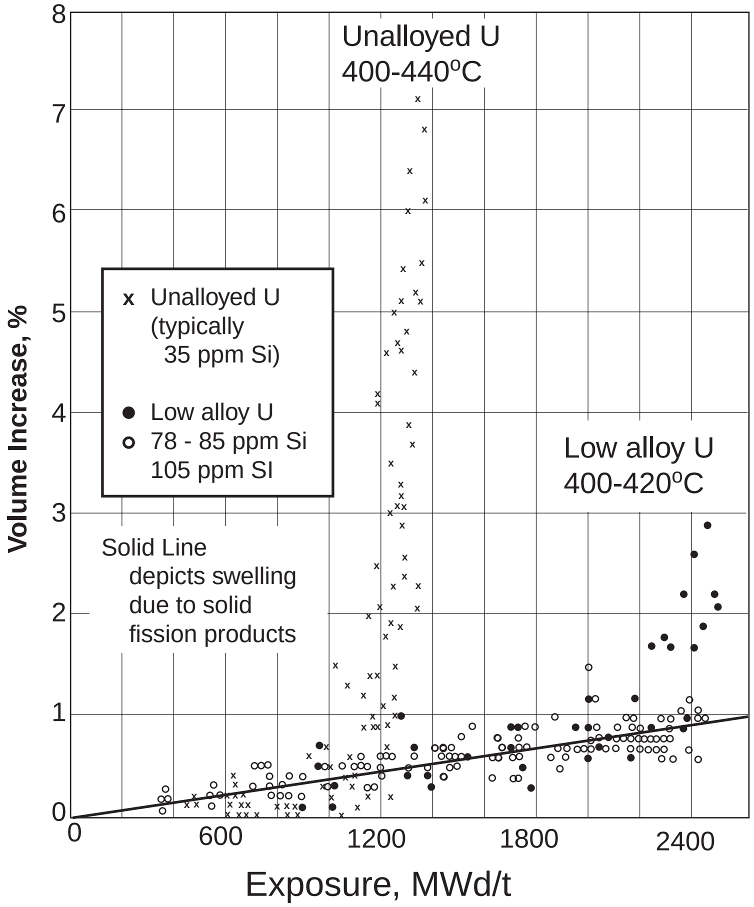

Figure 3. Swelling of unalloyed uranium and low silicon-containing alloy fuel elements during irradiation

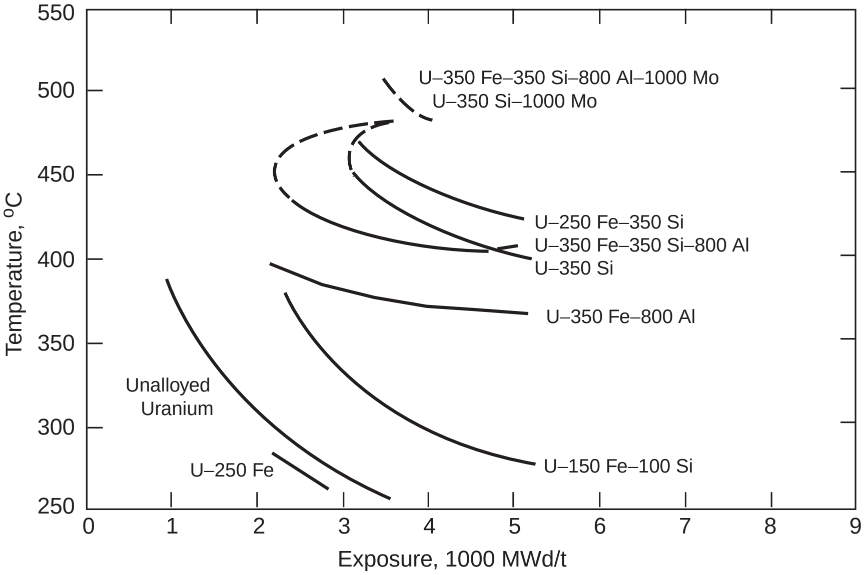

Figure 4. Temperature-exposure thresholds for swelling of uranium fuel (alloying concentrations in parts per million)

The principal features that emerged from these investigations were as follows:

- Unalloyed uranium swelled at relatively low temperatures and exposures, depending in some measure on impurity (especially silicon) concentrations in the metal.
- Alloying additions of low-solubility elements increased swelling resistance and allowed higher temperatures and/or longer exposures during irradiation before swelling began. Silicon concentrations of 250 to 350 ppm alone, or in conjunction with other alloying elements, especially molybdenum, were most effective for exposures up to 5000 MWd/t. Aluminum and larger silicon additions were less effective, and incremental iron or chromium additions produced no discernible benefit. At exposures over 10,000 MWd/t, alloys containing 800 ppm aluminum in conjunction with other alloying elements were more swelling-resistant than those containing 250 to 350 ppm silicon.
- Irradiation produced gross distortions of grain structure and alloy-phase distributions that minimized the effects of the initial structures. The grain distortions resulted from interactions of individual grains undergoing anisotropic growth during irradiation. The cavities formed as a consequence appeared more closely related to the characteristics of the substructure that emerged during irradiation than to the original grain structure, which was essentially destroyed. The alloy content rather than the grain structure appeared to be the primary controlling factor in swelling resistance. Alloy phases initially present in the structure, such as UAl_2, were dispersed and possibly dissolved during irradiation, and the relative swelling resistance of the alloys appeared to be established by the characteristics of the submicroscopic dispersions of alloy phases thus formed.

The above results served for many years to define limiting conditions for using metallic uranium fuel in SRP reactors without detrimental swelling. Tubular elements irradiated to high exposures occasionally exhibited gross core cracks, but such cracks did not penetrate the aluminum cladding unless swelling thresholds were exceeded.

Extensive studies of the fundamentals of swelling of uranium and uranium alloys pursued at the Savannah River Laboratory and at Hanford indicated that cavitational swelling, closely related to anisotropic growth in uranium, probably represented the first manifestation of the later generally recognized problem of void formation induced by fast-particle irradiation of many materials (McDonell 1973).

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End Notes

1. Metallic uranium exhibits three crystallographic structures depending on temperature (Foote 1956). Up to 668°C, the metal has an orthorhombic crystal structure (alpha phase); from 668-774°C, a tetragonal crystal structure (beta phase); and above 774°C to the melting point 1132°C, a cubic crystal structure (gamma phase). The orthorhombic alpha phase is highly anisotropic exhibiting in single crystals marked differences in properties and irradiation growth in different crystallographic directions. Polycrystalline uranium specimens share the anisotropic properties and behavior of the single crystal, to the extent the crystals are preferentially oriented in the specimens. Uranium specimens with preferred crystallographic orientations are said to be "textured".

Biographies

William R. McDonell

B.S., Chemistry, University of Michigan, 1947; M.S., Chemistry, University of Michigan, 1948; Ph.D., University of California Berkeley, Nuclear Chemistry, 1951.

Employed E.I. duPont de Nemours & Co.-Argonne National Lab and Savannah River Plant, 1951-89 (Research Associate).

Westinghouse Savannah River Co. 1989-95 (Advisory Scientist).

Tetra Tech NUS Co., 1995-96, 98-present (Environmental Scientist).

Participated in the development of uranium fuel elements for Savannah River reactors, product forms for Pu +238, Cm −244, and Cf −252 radioisotopic heat and radiation sources, technology and costs of disposal of high-level, transuranium, and low-level radioactive wastes. Principal research interests in effects of radiation on metals, ceramics, and glasses. Recent work devoted to long-range planning for disposition of spent reactor fuels and radioactive materials.

George R. Caskey, Jr.

B.S., Metallurgical Engineering, University of Illinois, 1950; M.S., Metallurgy, MIT, 1952; Ph.D., Materials Science, MIT 1969.

Employed E.I. duPont de Nemours & Co. 1952-1989 (research staff metallurgist); Virginia Polytechnic Institute 1981 (visiting professor); Westinghouse Savannah River Co., 1989-1993 (fellow scientist); Oak Ridge Institute for Science and Education, 1997-present (scientist emeritus).

Participated in development of fuel elements for the Savannah River reactors, compatibility of materials with product streams for the processing and storage of hydrogen isotopes, evaluation of the effects of irradiation and stress corrosion cracking on aging of reactor components and reactor service life management, and technology in support of storage of spent nuclear fuel. Research interests focused on austenitic stainless steels, irradiation effects, stress corrosion cracking, and compatibility with hydrogen isotopes.

Carl L. Angerman

B.S., Chemistry, Bowling Green State University, 1952.

Employed E.I. duPont de Nemours & Co.-Argonne National Laboratory, 1952-53, and Savannah River Plant, 1953-89 (research associate), Westinghouse Savannah River Co., 1989-91 (advisory scientist).

Participated in development of fuel elements and structural material for Savannah River reactors. Principal research interests in applications of optical and electron metallography to fabrication and irradiation behavior of uranium and aluminum-based reactor fuel alloys, selection of superalloys for encapsulation of Co-60 heat sources, and effects of tritium exposure on mechanical properties of stainless steel containment materials.

Aluminum-Lithium Technology and Savannah River's Contribution to Understanding Hydrogen Effects in Metals

M. R. (Mac) Louthan, Jr.

Abstract

Exposure of aluminum-lithium alloy targets to thermal neutrons in Savannah River Site (SRS) reactors produced tritium, the radioactive isotope of hydrogen that is used in nuclear weapons. The target development program assessed the metallurgical and mechanical properties of aluminum-lithium alloys and determined the factors effecting tritium retention and migration in the target material. An understanding of hydrogen behavior in the target and in other tritium-containing materials was required to assure that tritium-handling operations at SRS were both safe and efficient. The aluminum-lithium and hydrogen-in-metals studies provided the technical basis to assure the combination of tritium retention in target materials during irradiation and transport, successful tritium removal by processing targets in the extraction facility, and successful operation of a tritium packaging and handling systems. The hydrogen-in-metals research programs, which were focused on plant operating needs, also led to discoveries that influenced numerous technologies not generally associated with weapons-materials production. Experimental studies of hydrogen in zirconium and titanium alloys provided a technical basis to avoid stress orientation of hydrides in nuclear fuel claddings and delayed failure of welds on titanium airframes. The susceptibility of austenitic stainless steels to hydrogen embrittlement was demonstrated and broad-based models for hydrogen embrittlement processes were developed. Measurement and analysis of hydrogen uptake and migration kinetics in a variety of engineering materials provided the basis to demonstrate hydrogen transport by dislocations, to identify short circuit diffusion paths in multiphase alloys, and to understand hydrogen trapping at extraordinary sites in a metal lattice. Hydrogen solubility and diffusivity equations were also established for a number of metals. This paper highlights some of the key discoveries associated with the aluminum-lithium and hydrogen-in-metals studies at SRTC.

Introduction

Tritium, an isotope of hydrogen, is a critical component in the construction of thermonuclear weapons. There is very little tritium existing in nature, thus, to support U.S. weapons programs, tritium was produced by the irradiation of lithium in nuclear reactors at SRS. The successful use of tritium to increase the yield of nuclear weapons required a detailed understanding of the behavior of tritium production, extraction, and containment systems. This paper highlights the aluminum-lithium and other hydrogen in metals technologies that emerged from Savannah River during the past four decades of tritium production.

The isotopes of hydrogen are protium, deuterium, and tritium. The nuclei of these isotopes contain one proton and either zero (protium), one (deuterium), or two (tritium) neutrons. All three of the isotopes occur in nature, but most of the existing tritium has been prepared artificially through nuclear reactions such as:

$$_3\mathrm{Li}^6 + {}_0\mathrm{n}^1 = {}_1\mathrm{H}^3 + {}_2\mathrm{He}^4.$$

This reaction, which depicts the lithium isotope of mass six absorbing a neutron (mass one) and decaying into tritium (mass three) and helium (mass four) atoms, was the basis for tritium production at SRS. Development of the technologies required to safely perform this reaction in a nuclear reactor and to process, contain, and package the resulting tritium was a major SRS contribution to the success of the U. S. nuclear weapons program.

Tritium produced at SRS was mixed with deuterium for use in a nuclear weapon. Such tritium-deuterium mixtures are used to boost the yield of nuclear weapons through thermonuclear reactions such as

$$_1H^3 + {}_1H^2 = {}_2He^4 + {}_0n^1 + 18\ \text{MeV}.$$

The amount of energy released by the fusion of deuterium and tritium atoms to form a helium atom and a neutron is over four million times as great as the amount of energy released by chemical reactions such as

$$C + O_2 = CO_2.$$

The great amount of energy released by the thermonuclear reaction is a primary reason that nuclear weapons can be engineered to have multi-megaton yields. Successfully using tritium to boost the yield of nuclear weapons requires a detailed understanding of the behavior of tritium in the production, extraction, containment, and packaging systems. Tritium must be carefully contained and handled at all stages of processing because it undergoes radioactive decay by emission of a β-particle (an electron) and conversion to a helium atom of mass three.

The metallurgical characterizations of aluminum-lithium alloys at Savannah River, supplemented by national laboratory investigations, provided the technical basis for successful design, fabrication, and irradiation of tritium production targets. This work was one of the first major efforts to fabricate and use standard components manufactured from aluminum-lithium alloys. Many of the property measurements made during the characterization still serve as benchmark references for the aluminum-lithium system. Additionally, the Savannah River efforts to establish foundry practices, to develop fabrication techniques and heat treatment schedules, and to understand material behavior provided part of the experience base required for the aluminum-lithium industry. This industry is now providing aluminum-lithium alloys for aircraft and aerospace components, partially because of the technology and experience developed for tritium production targets.

The broad-based impact of tritium target technology development is even more apparent when the associated hydrogen-in-metals technologies are considered. An understanding of tritium, or hydrogen, uptake and diffusion in metals was required to assure tritium retention in the target during irradiation and storage and tritium release during extraction operations. An understanding of the behavior of hydrogen, including hydrogen effects on the mechanical properties of structural and containment materials, was required for safe and efficient operation of the tritium extraction, packaging, and storage systems. This requirement provided a gateway to large-scale hydrogen-in-metals research because the expertise and facilities necessary for successful tritium operations are identical to those required for solving many other hydrogen-in-metals problems.

Aluminum-Lithium Alloys

Lithium was alloyed with aluminum and processed through the Savannah River fuel and target fabrication facility to form a tritium production target. The targets were then exposed to neutrons (irradiated) in the Savannah River reactors. The irradiated targets were removed from the reactor, allowed to cool in large pools or basins, and ultimately transferred to the extraction facility where tritium was removed, purified, stored, and packaged.

Lithium reacts so readily with oxygen and moisture that it is used as a scavenging/purifying agent for inert gases. This high chemical reactivity prevents the direct exposure of lithium to either air or water. The SRS reactors used heavy water (deuterium oxide) to cool the fuel and target elements and to moderate (slow) the neutrons produced in the fuel elements. In addition, irradiated components are cooled in water-filled storage basins. Therefore, unprotected lithium could not be used as a target material for tritium production. Aluminum was the material selected to protect the lithium and to contain any tritium produced in the target element.

Two protection and containment barriers were established during the fabrication of the aluminum-lithium, tritium production targets. The first barrier against lithium interactions with, or tritium release to, the surrounding environment was the aluminum matrix that surrounded lithium-rich particles in an aluminum-lithium alloy. Predetermined quantities of lithium were placed in an already molten aluminum bath to form this alloy. The melt was then cast to form right circular cylinders of various lengths. These cylinders were covered with relatively pure aluminum and further treated to form slugs that were 2 to 3 centimeters in diameter or tubular elements that were either 4 or 9 centimeters in diameter. The aluminum coverings or claddings for the aluminum-lithium alloys were approximately 0.75 millimeters thick and provided the second protection/containment barrier.

Thousands of target tubes were needed to meet tritium production requirements. Fabrication techniques, alloy specifications, inspection procedures, and irradiation conditions were chosen to assure:

- Reproducible target production to very exacting specifications
- Cladding integrity throughout fabrication, irradiation, and storage
- Tritium containment during irradiation and storage
- Dimensional stability during irradiation
- Efficient tritium release during extraction.

Effective standardization of the processes associated with target fabrication and tritium production required an understanding of the physical and mechanical properties of aluminum-lithium alloys.

Microstructure controls the properties of metals and alloys. Most engineering alloys contain many, very small crystals of various orientations. The individual crystals are termed grains and the size, shape, and arrangement of the grains is part of the materials microstructure. Microstructure is revealed by the examination of specially prepared samples. This examination is termed metallography and generally requires the use of a microscope to reveal the grain structure. The boundaries between grains, precipitates, and inclusions are other examples of microstructural elements in metals. The number, size, shape, and distribution of the microstructural elements can be controlled through metallurgical practices such as heat treatment and deformation. Different production practices will produce different microstructures in the same metal or alloy. Because microstructure plays a major role in controlling the properties of metals and alloys, the properties of a given alloy are significantly influenced by the production practice. The aluminum-lithium alloys used for the tritium production targets contained microstructures that had to be characterized to assure successful production practices. The effects of foundry and fabrication practices on microstructure were determined and techniques to control the microstructure were established. The effects of grain size and precipitate morphology on the mechanical properties were found to be of particular importance.

The metallurgical characterization of precipitate behavior in the target alloys required knowledge of the solubility of lithium in aluminum. The temperature dependence of lithium solubility in aluminum was measured in the early 1960s (Costas and Marshall 1962). This work provided a partial basis for alloy selection and heat treatment practices that increased the room temperature yield strength of aluminum from slightly less than 50 MPa to over 250 MPa. The alloy strength increased with increasing lithium, up to approximately 5 wt% Li, and could be modified through heat treatment. Test samples were extruded and aged (held at an elevated temperature for a prescribed period of time) to develop the high strengths.

Irradiation generates heat at the target interior because of the energy deposition associated with neutron absorption and because of the energy released by the transmutation of lithium to tritium and helium. The heat is removed from the target by reactor coolant flowing across the cladding. This produces a thermal gradient in the target. The size of the gradient and the maximum internal target temperature depend on the amount of heat generated, the temperature of the coolant, the thermal conductivity of both the aluminum-lithium alloy target and the cladding material, and the size of the target. Therefore, the thermal conductivity of the target material needed to be established, and the effects of lithium content and irradiation on thermal conductivity were determined. It was discovered that the temperature gradients in a target during irradiation caused lithium to migrate from hot regions to cold regions (Costas 1962). The rate of migration was dependent on lithium content, temperature, and temperature gradient. Migration was negligible if the temperature was below 473 degrees K or if the lithium content was below 1.5 wt.%. Furthermore, the tendency for lithium migration was also affected by the metallurgical condition of the target material. These were significant observations because lithium migration, which could distort the target, disrupt the coolant flow, and adversely affect reactor operations, could be controlled through alloy selection, heat treatment, and target design.

The effects of alloy selection, fabrication practices, and heat treatment on the mechanical properties of the aluminum-lithium alloy were determined. Tensile properties, hardness, and creep rates were measured, and this knowledge was integrated into the target development processes. Creep, or time-dependent plastic deformation, was of concern because many aluminum alloys tend to creep at temperatures below the anticipated reactor operating temperature. Creep can be caused by atom migration and, in the target alloy, could be controlled by migration of aluminum atoms, lithium atoms, or both. Atom migration, or diffusion, in materials is a thermally activated process that is typically described in terms of the diffusivity. The diffusivity of lithium in aluminum was measured at Savannah River (Costas 1963). The similarity between the temperature dependence for lithium diffusion and for creep in aluminum-lithium alloys demonstrated that lithium atom movement controlled creep. This conclusion was consistent with the observation that, during creep tests, lithium diffused to and precipitated as LiAl on grain boundaries oriented 45-90 degrees to the stress axis (Marshall 1961). This is an example of how microstructure can change when an alloy is exposed to a new environment. The selective precipitation occurred because the density of the LiAl phase, 1.75 gm cm^{-3}, is less than the density of aluminum-lithium matrix, thus the preferential precipitation of LiAl on favorably oriented grain boundaries would selectively expand the alloy along the stress axis. However, lithium redistribution was not observed in tests at temperatures below approximately 473 degrees K, thus providing additional evidence of the importance of temperature control to target stability.

Tritium and Helium in the Target

Studies at Savannah River demonstrated that irradiation changed the metallurgical condition and microstructure of the tritium production targets. Neutron absorption caused the aluminum and lithium atoms in the target to be displaced from their equilibrium positions—termed displacement damage—and produced transmutation products such as tritium, helium, and silicon. Tritium and helium were produced from lithium-6 (Li-6) atoms, and silicon was produced from aluminum. The displacement damage increased the number of disruptions in the alloy, thus increasing the strength and decreasing the ductility and toughness of the material. Silicon accumulations had a similar effect. The level of change in the mechanical properties of the target materials increased with increasing neutron fluence (number of neutrons hitting the target). The amount of tritium and helium generated in the target also increased with increasing neutron fluence. Tritium and helium are relatively insoluble in aluminum and, given the opportunity, agglomerate and precipitate to form gas bubbles in the aluminum alloy matrix. The nucleation and growth of gas bubbles could cause the target to swell during irradiation in much the same fashion as bread rises during baking. Excessive swelling impairs target performance by inhibiting coolant flow, increasing the target temperature, and compromising tritium containment.

The tritium and helium formed in the target behave somewhat differently. The newly formed helium atoms are primarily located in the aluminum matrix while the tritium atoms are distributed in both the matrix and the lithium-rich LiAl phase (Owen and Randall 1976). The tritium atoms react with the LiAl to form lithium tritide and free aluminum by the chemical reaction

$$\mathrm{LiAl} + \mathrm{T} = \mathrm{LiT} + \mathrm{Al}.$$

The equilibrium tritium pressure associated with this reaction is temperature dependent and is given by (Peacock et al. 1995)

$$p_{(equi)} = 351 \exp(-10{,}700/RT) \text{ atm}.$$

This pressure represents the tritium pressure inside a gas bubble or cavity in the target material and, at a reactor operating temperature of 373 degrees K, is only 0.0002 atm if lithium atoms are available for interaction with the newly formed tritium. This pressure is too low to cause target swelling. Additionally, this very low effective pressure provides a minimal driving force for tritium release and thus enhances tritium retention in the target. In the absence of lithium atoms for the reaction, the effective tritium pressure would be given by the relationship

$$p^{1/2} = C/[0.00034 \exp(-15100/RT)] \text{ atm}^{1/2}$$

where C is the tritium concentration, expressed in parts per million (Louthan et al. 1976a). The pressure calculated from this relationship when C is 10 parts per million could not be contained if gas bubbles developed in the target. Therefore, lithium must remain in the target alloy throughout the irradiation in order to prevent tritium-induced swelling and tritium release.

These observations provided the technical basis to specify a maximum Li-6 enrichment of 50%. The Li-7 atoms are not involved in the transmutation reaction and therefore remain in the aluminum-lithium alloy throughout irradiation. This specification assured that even if the target remained in the reactor until all the Li-6 was transmuted to tritium and helium, there would be enough Li-7 remaining in the aluminum-lithium target material to trap (react with) any tritium produced in the target. In addition to preventing tritium induced swelling, trapping greatly reduced tritium migration rates and prevented diffusion-induced tritium losses when irradiating, storing, and transporting targets.

Helium is an inert gas and generally does not interact with other atoms or molecules. Therefore, chemical reactions, similar to the tritium trapping reaction, could not be used to control helium-driven swelling of irradiated targets. This fact, coupled with the success of tritium trapping in preventing tritium-driven swelling, demonstrated that swelling under irradiation is primarily due to helium generation (McDonnell 1989). One helium atom is produced for every tritium atom created by the $^{6}\mathrm{Li}(n,\alpha)^{3}\mathrm{H}$ reaction. Agglomeration of the helium atoms into large gas bubbles would compromise target performance by causing large-scale swelling. Fortunately, the helium atom size is such that, at low helium concentrations, the helium atoms are trapped at microstructural discontinuities in the aluminum-lithium alloy. This type of trapping leads to the formation of small clusters of helium atoms and does not cause appreciable swelling. However, the cluster size grows with irradiation exposure and eventually the build-in of helium will cause the target to swell. The helium concentration required for the onset of swelling is sensitive to the temperature and decreases as the temperature increases.

The swelling threshold at SRS reactor operating temperatures occurs at a helium content of approximately 0.067 wt %. Although this weight fraction looks small, the helium contained in 1 cc of aluminum-lithium alloy at the swelling threshold would occupy approximately 100 cc if released into the air at room temperature. Compression of the 100 cc of helium into a 1-cc volume requires a pressure of 1470 psi. A higher pressure is required to contain the helium in a volume smaller than 1 cc. For example, a pressure of 14,700 psi is required to compress helium into a gas volume of 0.1 cc or 10% of volume occupied by the aluminum-lithium alloy that originally contained the helium. If the temperature is increased from room temperature to reactor operating temperatures for the target, the pressure exceeds 18,000 psi. If only 1% swelling is allowed, the helium pressure at reactor operating temperatures exceeds 180,000 psi. The yield strength of a non-irradiated Al-2wt%Li alloy at 373 degrees K is approximately 14,000 psi and an Al-4wt%Li alloy yields at a stress of approximately 23,000 psi.

The swelling comparisons illustrate the importance of limiting the amount of helium produced in an aluminum-lithium alloy target and demonstrate the necessity to control the chemistry of the target alloy. Integration of the SRS experience base with the accompanying increases in understanding of swelling processes provided the technology to develop the target irradiation limits that precluded excessive swelling during reactor operations.

Extraction of Tritium

The irradiated tritium production targets were removed from the reactor and stored in basins that contain about a million gallons of cooling water. The water flowed around, and through, the tubular targets. Water flow is driven by natural convection and maintains the target temperatures below those experienced during irradiation. Decay of the short-lived transmutation products and activated species lowers the heat-generating capacity of the target tubes, and, after several weeks of basin storage, the targets could be safely transferred to the tritium extraction facility. The entire removal and transfer operations involved remote operations through shielded facilities to minimize personnel exposure and assure operational safety.

Tritium was recovered by placing the irradiated targets in a stainless steel crucible, lowering the crucible-target assembly into an extraction furnace, evacuating the furnace chamber to a pressure below the decomposition pressure of lithium tritide, and heating the evacuated system to promote tritium release. Experimental measurements demonstrated that target swelling and helium release preceded tritium recovery (McDonnell 1989). Target heating promoted the agglomeration of the helium gas bubbles and the formation of interconnected porosity throughout the irradiated aluminum-lithium alloy. This agglomeration caused the target to swell, thus stretching the target cladding. The aluminum cladding then ruptured because its ductility had been reduced significantly by irradiation. Rupture of the cladding exposed the interconnected porosity and released the helium to the vacuum extraction environment. The low extraction pressure caused the lithium hydride, which was surrounded by aluminum, to decompose and release tritium to the extraction furnace. Virtually all the tritium produced during the reactor operation was removed from the target during the extraction operation.

The tritium and helium removed from the target during the extraction operation was collected in stainless steel tanks. The helium is not used in nuclear weapons; thus separation of the tritium from the helium is necessary. To separate the helium from the tritium, the collected gases were passed through a palladium diffuser. The diffuser is essentially a long, thin-walled tube. The gas mixture is passed on one side of the tube while a low pressure is maintained on the other side. The diffuser is heated to promote tritium uptake and diffusion in the tube wall. The combination of uptake and diffusion is termed permeation, and tritium and the other hydrogen isotopes will permeate (pass through) the tube wall. This allows for the collection of tritium, and any other hydrogen isotopes that may be present in the gas stream, on the evacuated side of the diffuser. Helium is virtually insoluble in most metals and alloys, including palladium, and does not permeate the tube. Diffuser operation thus provides a technique to separate the tritium (and other isotopes of hydrogen) from the helium in the extracted gas stream. However, all three hydrogen isotopes permeate the diffuser and the collected "tritium" gas stream also contains protium and deuterium. Protium, deuterium, and tritium separated from each other by cryogenic distillation and the final product is high-purity tritium. The high-purity tritium is packaged and stored for use in atomic weapons.

Associated Hydrogen-in-Metals Technologies

Advances in aluminum-lithium technology for tritium production at the Savannah River Site required the development of a large number of associated hydrogen-in-metals technologies. An understanding of tritium, or hydrogen, uptake and diffusion in metals was required to assure tritium retention in the target during irradiation and storage and tritium release during the extraction operation. The behavior of hydrogen, including hydrogen effects on the mechanical properties, in structural and containment materials was required for safe and efficient operation of the extraction, packaging, and storage systems. The expertise and facilities required for successful tritium production operations are identical to those required for solving most other hydrogen-in-metals problems. Therefore, the technical support system for the aluminum-lithium technology program also enhanced a variety of other hydrogen technologies not directly related to tritium production. The Savannah River work contributed significantly to the:

- Prevention of hydride-induced failure of zirconium alloys used in the nuclear power industry
- Avoidance of cracking during welding of titanium alloys of interest to NASA for supersonic aircraft
- Determinations of hydrogen solubility, diffusivity, and permeability in, and measurement of the effect of hydrogen on the mechanical properties of, infrastructure materials required to support a hydrogen economy
- Development of the fundamental aspects of hydrogen-metal interactions in a variety of metals and alloys
- Demonstration that helium, introduced through radioactive decay of tritium, increased the strength and decreased the ductility and weldability of tritium exposed metals
- Determination of the effect of helium on hydrogen-induced slow crack growth in austenitic stainless steels
- Discovery that helium implanted during irradiation can cause cracking during weld repair of nuclear reactor components.

The last three items listed have direct application to the selection and use of metals and alloys in fusion reactors and in advanced spallation neutron sources.

Hydride Cracking of Zirconium Alloys

Zirconium-based alloys are used for fuel cladding, process tubes, and structural elements in the nuclear power industry. These alloys have a strong affinity for hydrogen and, when sufficient hydrogen is absorbed, will precipitate hydride phases throughout the zirconium microstructure. These hydride precipitates can have deleterious effects on the mechanical properties of zirconium alloys. In the early 1960s, emerging experimental data suggested that the tolerance of zirconium alloys for hydrides was rather high and early concern over the potential for hydrogen embrittlement was subsiding. However, experiments at SRS demonstrated that the influence of hydrogen on the mechanical properties of Zircaloy (a zirconium-based alloy that contains small amounts of tin, iron, and nickel) was determined by the orientation of the hydride platelets. Small amounts of hydrides could have very deleterious effects on the mechanical properties if the platelets were oriented perpendicular to the direction of applied stress (Caskey et al. 1961; Louthan and Marshall 1963; Marshall and Louthan 1963). The orientation of the platelets was controlled by stresses in the Zircaloy during hydride precipitation. Platelets tended to precipitate with their broad faces perpendicular to tensile stresses and parallel to compressive stresses. This tendency was termed stress orientation (Marshall and Louthan 1963).

Zirconium alloys have a hexagonal-close-packed (hcp) crystal structure. Plastic deformation in hcp structures is anisotropic. This anisotropy leads to the development of crystalline textures or preferred orientations in most wrought products. The nature of the preferred orientation in any given component depends on the fabrication process used to manufacture that component. Measurements of the extent of stress orientation in tube sections with various preferred orientations (Marshall 1967) confirmed that the orientation of the hydrides in Zircaloy, and the susceptibility of Zircaloy to stress orientation were controlled by the preferred orientation of the Zircaloy matrix. Therefore, the processing technique use to manufacture the Zircaloy cladding, tube, or component had a major influence on the tendency of the materials to undergo stress orientation (Louthan and Marshall 1963; Marshall 1967). This observation demonstrated that a potentially significant problem with the use of Zircaloy as a nuclear fuel cladding could be mitigated through the selection of manufacturing technologies to control the preferred orientation of the material. These results provided the foundation for the technical basis that currently assures against adverse hydride orientations in fuel claddings and process tubing used in commercial power reactors.

Hydrogen in Titanium Alloys

The Savannah River observations that lithium diffused to and precipitated as LiAl on grain boundaries oriented 45 to 90 degrees to a tensile stress, and stresses caused preferential hydride orientations in Zircaloy led to the discovery that titanium alloys were also susceptible to stress orientation of hydrides (Louthan 1963). This observation was one of the keys to understanding delayed failure in titanium alloys.

Hydrogen uptake may occur in service or be introduced by fabrication processes such as welding. Titanium alloys react with moisture in the environment and release atomic hydrogen at the metal surface. This hydrogen may move through (permeate) the titanium and accumulate in regions of high tensile stress. The high stresses may be the result of service loads or may be residual stresses introduced by fabrication and/or assembly processes. When sufficient hydrogen has accumulated in the high stress area, a hydride phase will precipitate and will be oriented so that the broad face of the platelet is perpendicular to the tensile stress. This orientation is favorable for crack nucleation and/or growth along the hydride/metal interface or through the hydride itself. Once the hydride has cracked, the high stresses, which promoted hydrogen accumulation, will now be just beyond the crack tip. The accumulation, precipitation, cracking sequence will then be repeated.

The hydrogen accumulation and hydride precipitation processes are both diffusion controlled and require the passage of time. Under these conditions, hydrogen-induced cracking may initiate sometime after a crack-free part has been placed in service. The delay time for cracking can vary from hours to years, depending on the hydrogen content, stress level, microstructure of the titanium alloy, and temperature of exposure. The publication of observations on stress orientation of hydrides in titanium caused multiple interactions with National Aeronautics and Space Administration (NASA) because of the use of titanium in airplanes and aerospace applications. The continued importance of these results is apparent in the 1993 edition of the *Metals Handbook* (Metals Handbook 1993), which describes hydrogen-induced delayed cracking of titanium welds as resulting from the time-dependent precipitation of stress oriented hydrides in the high tensile stress regions of the welds.

The National Aeronautics and Space Administration sponsored several experimental programs at Savannah River, including an investigation of hot salt stress corrosion cracking of titanium alloys. The research, conducted in the mid to late 1960s, demonstrated the hot salt stress corrosion cracking process involved two distinct steps: crack initiation and crack propagation (Rideout et al. 1969; Rideout et al. 1970). Crack initiation occurs after an incubation period that depends on the temperature of exposure, chemical composition of the salt deposit, and composition of the titanium alloy. The presence of moisture in the salt deposit and the formation of HCl gas by hydrolysis also play vital roles in the cracking sequence. Radiotracer studies, using tritium as a tracer, showed that corrosion produced hydrogen is absorbed by the metal. It was proposed that the absorbed hydrogen accumulated in regions of high, localized tensile stresses. Cracks initiated when the hydrogen concentration exceeded some critical level. Crack propagation was found to be less sensitive to temperature than crack initiation. The stress level and the nature of the salt deposit controlled the rate of crack propagation. Crack propagation occurred primarily by mechanical rupture processes and little or no evidence of corrosion was found on the fracture surfaces. The importance of these observations to understanding and mitigating hot-salt stress corrosion cracking in titanium alloys was emphasized in an article published in the *ASM Source Book on Titanium and Titanium Alloys* (Petersen 1982).

Hydrogen Compatibility

The "Hydrogen Economy" emerged in the 1970s as a technology of importance to the future. This emergence was primarily because of the long lines at gas pumps but also as part of the ongoing quest for clean energy technologies to replace burning fossil fuels. The Energy Research and Development Administration (ERDA), the predecessor organization to the Department of Energy, established an Interagency Agreement through which NASA provided assistance to ERDA's hydrogen energy storage program. The production, storage, transportation, and use of hydrogen as an energy carrier were investigated through this cooperative research and development program and through associated programs conducted at various ERDA sites, such as Savannah River. The Savannah River contributions to the "Hydrogen Economy" technologies were determinations of the compatibility of metals and alloys with hydrogen environments.

Compatibility studies included the determination of hydrogen uptake and migration kinetics in, and measurements of hydrogen effects on the mechanical properties of, structural materials. The Savannah River hydrogen-in-metals technologies, developed to support the production, extraction, storage, and packaging of hydrogen isotopes for nuclear weapons were identical to the hydrogen-in-metals technologies required to support the hydrogen economy.

Data were necessary to assure satisfactory hydrogen containment in hydrogen storage vessels, gas transfer lines, valves, pumps, and the associated storage and transfer equipment. Satisfactory containment included assuring that potential losses of hydrogen because of uptake and/or permeation were minimized and that the materials of construction were compatible with high-pressure hydrogen environments. Savannah River had investigated the behavior of approximately 50 commercial alloys in hydrogen environments by the mid 1970s (Louthan and Caskey 1976a).

Radioactivity provides a significant tool that enhances the detectability of tritium. This enhanced detectability allowed the measurement of the behavior of hydrogen in metals under conditions not previously accessible to measurement. The diffusivity and permeability of hydrogen in cobalt (Caskey et al. 1974), nickel (Louthan et al. 1975a), molybdenum (Caskey 1975), copper (Caskey et al. 1976), aluminum (Louthan et al. 1976a), and titanium alloys (Louthan et al. 1975b) were measured at near ambient temperatures. Hydrogen diffusivity, solubility, and permeability were also measured in austenitic and ferritic/martensitic steels (Louthan and Caskey 1976; Louthan and Derrick 1975; Caskey and Derrick 1974; Louthan et al. 1976b). The data and analysis from the studies provided the technical basis to assess hydrogen uptake and migration in many of the commercial alloys that were being considered for use in the hydrogen production, storage, and distribution systems required if the U.S. were to move toward a "hydrogen economy". The studies demonstrated that hydrogen trapping at metallurgical discontinuities (dislocations, grain boundaries, phase boundaries, alloy and impurity atoms, etc.) played a major role in hydrogen uptake and migration (Louthan et al. 1975a; Caskey et al. 1976; Caskey and Allinger 1974; Louthan 1974) and that surface films, normally present on these commercial alloys, play a major role in hydrogen absorption and permeation processes (Louthan and Caskey 1976; Louthan et al. 1975b; Louthan and Derrick 1975). Generally, the highest permeabilities to hydrogen were found in the iron-based alloys. The transition metals such as nickel and cobalt also showed high permeabilities to hydrogen. Aluminum and copper alloys showed the lowest permeabilities, partially because the hydrogen solubility in these metals is so low.

The room-temperature tensile properties of these same commercial alloys were measured before, during, and after exposure to high-pressure hydrogen environments (Louthan and Caskey 1976; Louthan 1974; Louthan et al. 1972; Capeletti and Louthan 1977). These studies demonstrated that all alloys show some evidence of susceptibility to hydrogen embrittlement. The aluminum alloys were the most resistant to hydrogen-induced degradation. Copper alloys also displayed excellent resistance to hydrogen damage. Iron-, nickel- and titanium-based alloys were all very susceptible to hydrogen damage; however, the extent of damage was dependent on pre-test exposure conditions, sample surface finish, hydrogen pressure during testing, and the metallurgical condition of the alloy. The data and analyses developed from these Savannah River investigations continue to provide a significant part of the technical basis for selection and use of alloys for service in hydrogen environments. This is particularly true for austenitic stainless steels because of a handbook published by Savannah River (Caskey 1983).

Fundamental Aspects of Hydrogen-Metal Interactions

The development of basic data necessary to assure the compatibility of metals and alloys with hydrogen was accompanied by an increased understanding of the fundamental aspects of hydrogen-metal interactions. The Savannah River data and analysis demonstrated that absorption and permeation of hydrogen, as well as the final distribution of hydrogen throughout a metal structure, could not be predicted accurately by the usual thermodynamic and diffusion relationships. Surface effects (particularly the properties and stability of oxide films), trapping by impurity and microstructural defects (discontinuities), and hydrogen transport by moving dislocations all profoundly influence hydrogen motion and distribution (Louthan and Caskey 1976). These three factors also effect hydrogen embrittlement because the quantity and distribution of absorbed hydrogen are directly related to the hydrogen embrittlement mechanism. Because of the importance of trapping to the understanding of hydrogen embrittlement processes, finite-difference techniques were developed to analyze the trapping process (Caskey and Pillinger 1975).

Tensile tests of stainless steel and nickel in high-pressure tritium environments (Louthan et al. 1972; Donovan 1976) demonstrated that plastic deformation had profound effects on the absorption and subsequent distribution of tritium in the test material. Localized, high tritium concentrations were identified by autoradiographic techniques (Louthan et al. 1972). Tensile and/or bend tests with tritium charged iron, Type 304L stainless steel, Alloy 718, and Type 5086 aluminum demonstrated the tritium was associated with dislocations and moved when the dislocations moved (Louthen et al. 1972; Donovan 1977). Dislocations are microstructural defects that are found in virtually all metals and alloys. Dislocation motion is generally responsible for plastic deformation in metallic structures. The observations that hydrogen attached to and moved with dislocations and that such motion can lead to localized, high hydrogen concentrations are two of the basic precepts for hydrogen embrittlement in metals and alloys that do not form hydrides or other hydrogen-rich phases. The Savannah River paper that initially presented these observations was republished in the ASM International book, *Hydrogen Damage*. This book collected, from the thousands of papers discussing hydrogen-in-metals, 30 "key contributions to the understanding of hydrogen damage and to our efforts to overcome the problems it continues to bring" (Hydrogen Damage 1975).

Autoradiographic studies demonstrated that hydrogen diffusion in rutile is anisotropic (Caskey 1974a). Rutile is titanium dioxide and forms naturally on the surface of titanium alloys exposed to air, water, and/or other oxygen containing environments. Oxide film formation on titanium is strongly influenced by the orientation of the underlying metal because the oxide forms in specific orientations with respect to the underlying metal. The oxide-metal epitaxy and the anisotropy in hydrogen diffusion were used to explain hydriding processes in titanium alloys used in hydrogen service (Caskey 1974b).

Other Savannah River studies related to fundamental aspects of hydrogen-in-metals include finding that:

- Isotopic effects on hydrogen transport in nickel are predictable from absolute rate theory if hydrogen acts as an anharmonic oscillator (Louthan et al. 1974a).
- Hydrogen expands the lattice and lowers the stacking fault energy of austenitic steels (Holzworth and Louthan 1968).
- Hydrogen lowers the cohesive energy of coherent twin and grain boundaries (Capeletti and Louthan 1977; Caskey 1983).
- The heat of solution for hydrogen is related to the electron density of states at the Fermi surface (Louthan et al. 1972).
- Hydrogen increases the lattice friction stress of austenitic stainless steels (Caskey 1983).
- Hydrogen can either suppress or enhance the formation of strain-induced martensite in austenitic stainless steels depending on the composition of the steel and temperature (Caskey 1983).
- High hydrogen solubility, low stacking fault energies, and high yield strength (Louthan et al. 1972) promote hydrogen embrittlement.

These fundamental observations provided a technical basis for a phenomenological model for hydrogen embrittlement (Louthan 1987).

Tritium Decay and Helium Accumulations

The radioactive decay of tritium is by the $^3H(b)^3He$ reaction. The helium (^3He), in a tritium-charged metal or alloy, is "born" in the metal lattice. Normally, helium does not dissolve in metals, but the in-lattice birth places a helium atom in solid solution in the metal and can lead to a significant helium build-in when tritium charged samples are stored for long periods of time. Helium atoms do not "fit" in either the normal lattice sites or the interstitial sites where hydrogen atoms generally reside. Helium embrittlement of metals is well known in the nuclear power industry because helium atoms are transmutation products in many nuclear reactions. This embrittlement is considered to be an elevated temperature process and was observed in tritium charged Type 309 stainless steel in the mid 1970s (Louthan et al. 1976c). These initial studies showed that helium build-in increased the strength of the steel but did not significantly effect the ductility until the helium-containing material was heated for a half-hour at 973oK. Subsequent studies demonstrated that elevated temperatures were not requiref for embritvlemgnt (Rawl et al. 1980; West and Rawl 1980). Austenitic stainless steel tensile bars that were tritium charged and aged until the helium concentration exceeded 200 appm and then tested at room temperature, failed along grain boundaries. The ductility was not restored when the samples were vacuum outgassed to remove the remaining tritium (West and Rawl 1980).

Room temperature helium embrittlement was not anticipated because helium is highly trapped. The elevated-temperature helium embrittlement studies suggested that the helium-induced failures result from the accumulction of helium bubbles along grain boundaries (Louthan 1976c). This failure mechanism requires helium migration to grain boundaries, which should be minimal at or near room temperature. Studies with iron and austenitic stainless steel confirmed the intergranular failure process and showed that the formation of helium bubbles was accelerated by a tensile stress (Donovan 1980). These observations demonstrated that helium-induced effects on the mechanical properties had to be evaluated before the long-term safety of tritium containment systems could be assured. Additionally, these results had significant implications to the weldability of irradiated metals and alloys.

These implications became apparent during attempts to repair the Savannah River C-Reactor tank by welding (Kanne 1988). Stress corrosion cracks in a curved transition piece that connected the tank sidewall to the bottom of C Reactor caused the reactor tank to leak in the late 1960s. The reactor was shut down, repaired (using remote gas tungsten arc welding techniques), and returned to service. The tank leaked again in 1984, and a program was initiated to repair the new leaks. Placement of patches over the cracks was, as in 1968, determined to be the best method of repair (Kanne 1988). A robotic-operated service arm was designed and built. Approximately 20 types of end effectors, including those for repair welding, were included in the design. A segmented patch was welded in place and then bubble tested by pressurizing the space between the tank wall and the patch with gas, raising the water level inside the tank, and monitoring for bubbles. Several leak sites were found. Subsequent, dye-penetrant testing showed toe cracking in the welds on the reactor wall (Kanne 1988). The tank wall contained approximately 3 appm of helium, which was introduced into the stainless steel by irradiation-induced transmutations of the alloy element nickel and the impurity element boron. An extensive test program demonstrated that the toe cracks were caused by the welding-induced agglomeration of helium bubbles along the grain boundaries. Test welds in materials that were charged with helium by the radioactive decay of absorbed tritium played an integral role in proving that the weld toe cracking was caused by the presence of helium (Kanne 1988).

Demonstration that small quantities of helium dramatically reduced the weldability of irradiated stainless steel had major implications on the design and repair of components and systems for reactors, accelerators, and other systems where helium implantation will accompany service. Additionally, the use of tritium charged and aged samples to simulate irradiation-induced helium became a standard technique to evaluate techniques for weld repair of irradiated materials. The 1988 discovery of helium effects on the weldability of metals and alloys provided the technical basis for several other Savannah River programs to validate weld repair techniques, quantify the level of helium required to cause weld cracking for different weld techniques, and model the weld cracking processes. One of the current programs is focussed on applications to fusion energy systems.

Hydrogen, Helium, and Slow Crack Growth

Tensile tests of hydrogen-charged austenitic stainless steels demonstrated that surface cracking accompanied plastic deformation. Metallographic examination of failed samples showed that hydrogen accumulation changed the fracture mode from a ductile to a brittle failure process. The brittle fracture modes included grain boundary and twin boundary cracking as well as cleavage fracture. The observation of hydrogen-induced brittle fracture modes raised concerns over hydrogen-induced slow crack growth in austenitic steel systems and components used for hydrogen containment (Caskey 1983). Slow crack growth could cause delayed failure hydrogen embrittlement and lead to sudden failure of in-service components. Tensile tests, which dominated the Savannah River hydrogen-in-metals studies throughout the 1970s, provided virtually no information that was relevant to slow crack growth. Therefore, by the mid 1980s, the hydrogen-in-metals test program had evolved to a focus on fracture mechanics studies to provide crack growth and fracture toughness data (Caskey 1983).

The fracture toughness of a material is generally expressed in terms of a stress intensity factor. This factor provides a measure of the level to which a crack or other sharp flaw enhances the effect of an applied stress. Values for the stress intensity factor depend on the type and magnitude of the load applied to the material and the size of the crack in the material. Failure takes place when the stress intensity factor reaches a critical level. This critical level is termed the fracture toughness of the material. Work at Savannah River demonstrated that tritium, and its decay product helium, lower the fracture toughness of austenitic stainless steels (Caskey 1983; Morgan and Tosten 1990). The extent of lowering depends on the strength (Caskey 1983) and metallurgical condition of the steel (Morgan and Tosten 1990). Other Savannah River studies demonstrated that hydrogen and helium could also cause slow crack growth in austenitic stainless steels that were loaded to stress intensities that were less than the critical stress intensity (Morgan and Lohmeier 1990). Subsequent, fracture mechanics type studies (Morgan and Tosten 1996) demonstrated that:

- Tritium exposed and aged steels had lower fracture-toughness values and less resistance to crack growth than unexposed steels.
- Fracture toughness values were reduced further as the concentration of helium increased.
- The tendency toward intergranular fracture increased as the concentration of helium increased.

Tensile testing demonstrated that, in addition to lowering the fracture toughness and the resistance to crack growth, both hydrogen and helium increased the strength and decreased the ductility of these steels (Morgan 1991). The helium-induced increases in strength enhance the effects of hydrogen on fracture toughness and crack growth. These data provided the technical basis to model slow crack growth in tritium containing stainless steels (Morgan 1997; Morgan 1999).

The model (Morgan 1999) shows that tritium is absorbed into the container wall and migrates through the wall by diffusion controlled processes. Helium then builds in from decay of the absorbed tritium. The helium build-in strengthens the metal but also lowers its resistance to crack growth. If the applied and residual stresses are high enough, a crack can nucleate and grow through the weakened or embrittled region. In this model, the crack could continue to grow in a stepwise fashion:

- Tritium diffusion and decay to helium embrittles the area near the crack tip.
- The crack then propagates through the embrittled zone.
- The crack arrests when it has propagated into non-embrittled material.
- Tritium absorption, diffusion, and decay begin to embrittle the new, near crack tip region.

As the crack grows through the material, the stress intensity level may increase (depending on the geometry and loading conditions). If this is the case, eventually, the non-embrittled region will be stressed to a level where crack growth can be driven by the presence of tritium. When this occurs, the material will fracture. This model—coupled with the Savannah River measurements of crack growth rates, critical stress intensity, factors and the effects of hydrogen and helium on critical stress intensity factors—supports the technical basis to assure the safety of tritium storage containers packaged and handled at Savannah River.

The observation that hydrogen and helium lower the fracture toughness and increase crack growth rates in stainless steels has significant implications to the behavior of metals and alloys in fusion and accelerator driven systems. The high-energy particle beams associated with these emerging energy systems will cause significant transmutations in the exposed areas. High concentrations of hydrogen and helium will therefore accumulate, decreasing the fracture toughness of the structural material. This decrease must be included in the design criteria for such systems.

Conclusion

The aluminum-lithium technology development at Savannah River, especially including hydrogen isotope and helium effects, has made significant scientific and technological contributions to the materials/metallurgical communities. These contributions extend far beyond the nuclear weapons materials production arena. In many respects, the contributions have favorably impacted the every day lives of most U.S. citizens and have the potential to continue to impact our society well into the 21st century.

Acknowledgment

The work described in this paper includes contributions from numerous engineers, scientists, and managers. The net result of these contributions is a chain of achievement linked together through the efforts of individuals. Unfortunately, listing all the individuals is impractical and listing only a few is unfair. Many of the technology developments sited were headquartered in materials oriented groups as the Savannah River Laboratory and the Savannah River Technology Center. The leadership of those groups has been outstanding and included: Phil Permar, Dick Huntoon, Jim Stone, Tami Capeletti, and Natraj Iyer. These leaders are thanked for providing a work atmosphere that fostered high-quality, cooperative research and development, and each individual contributor is thanked for being an integral part of that atmosphere and of the resulting accomplishments.

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Biography

M. R. (Mac) Louthan, Jr., earned a bachelor's degree in Metallurgical Engineering from Virginia Tech in 1960 and a Ph.D. in Metallurgical Engineering and Material Science from Notre Dame in 1967.

Mr. Louthan is a senior advisory engineer in the Materials Technology Section of Savannah River Technology Center. He is the author of approximately 200 technical publications, editor of 9 books, and a Fellow in ASM International. Mac developed the lecture "Why Stuff Falls Apart", which has been given over 200 times to colleges, universities, and civic and professional organizations. He has served as president of the International Metallographic Society, a member of the Board of Trustees of the National Youth Science Foundation, and chairman or co-chairman of 12 international conferences. He was a key reader for Metallurgical Transactions, a member of the editorial advisory board for Materials Characterization, and the series editor of Microstructural Science. Mr. Louthan has given invited presentations in throughout the U.S., Canada, Europe, and Asia, and is a member of Sigma Xi, Alpha Sigma Mu and Tau Beta Pi. Mac's awards include: the President's Award and two Best Paper Awards from the International Metallographic Society; the Instructor of Merit and the Distinguished Educator Awards from ASM International; an Award of Excellence from the Federal Laboratory Consortium for Technology Transfer; the Wine and two Sporn Awards for teaching excellence at Virginia Tech; and the Orth Award and three Westinghouse Signature Awards of Excellence from the Savannah River Site.

The Influence of Xenon-135 on Reactor Operation

Paul L. Roggenkamp

Abstract

Xenon-135 is a product of U-235 fission and has a very large neutron-capture cross section. It also decays radioactively with a half-life of 9.1 hours. Little of the Xe-135 results directly from fission; most comes from the decay chain, Te-135 (half-life, 0.5 min) to I-135 (half-life, 6.6 hr) to Xe-135. The combination of delayed generation and high neutron-capture cross section produces a diversity of impacts on nuclear reactor operation.

In the nuclear production reactors at the Savannah River Site (SRS), the flexibility in reactor charge design, the extensiveness of the reactor control system, and the multiplicity of monitoring systems for measuring the power distribution throughout the reactor enabled the effects of Xe-135 to be handled routinely. Occasionally, a Xe-135 disturbance arose that required special control measures, but for these also, the reactor systems were adequate.

This report describes measures that were necessary to handle the effects of Xe-135 in the day-to-day operations of power ascension, power level adjustment, reactor shutdown, and reactor restart. A few unusual but still significant cases of so-called "xenon oscillations" are discussed. The final section illustrates the effect of Xe-135 on the production process.

The time is Tuesday in late September 1944. The scene is the B Pile (graphite-moderated nuclear reactor) at the Hanford Reservation in Washington State. The dramatic discovery of the effect of Xe-135 is described this way (Rhodes 1986):

> The pile went critical at a few minutes past midnight; by 2 a.m. it was operating at a higher power level than any previous chain reaction. For the space of an hour all was well. Then Marshall remembers the operating engineers whispering to each other, adjusting control rods, whispering more urgently. 'Something was wrong. The pile reactivity was steadily decreasing with time; the control rods had to be withdrawn continuously from the pile to hold it at 100 megawatts. The time came when the rods were completely withdrawn. The reactor power began to drop down and down.'
>
> Early Wednesday evening B Pile died....
>
> Early Thursday morning the pile came back to life. It was running ... again. But twelve hours later it began another decline.

The culprit was Xe-135.

The solution for B Pile was to enlarge the core from the original 1500 tubes to 2004 tubes loaded with uranium. Provision had been made in the construction for such an eventuality.

Xenon-135, a fission product, has a neutron-capture cross section of 2,600,000 barns. In comparison, an atom of U-235 has a fission cross section of 550 barns. About 6.3% of fissions result in Xe-135, only 0.2% directly. The other 6.1% results from the radioactive decay of I-135, which has a half-life of 6.7 hours and a very small cross section for neutron capture. Xenon-135 decays radioactively with a half-life of 9.2 hours. This nuclide chain may be diagramed simply as in Table 1.

Under conditions of the operating Savannah River Site (SRS) nuclear reactors, Xe-135 is ten times more likely to be destroyed by neutron capture than by radioactive decay. These properties give Xe-135 its peculiar effects on reactor

Table 1. Nuclide chain

	Te-135 —>	I-135 —>	Xe-135 —>	Cs-135 —>	Ba-135
Half life	0.5 min	6.6 hr	9.1 hr	2.6 million yr	stable
Fission yield, %		6.1	0.2	0.1	

and reactor core design, operation (including oscillations in the spatial distribution of power), and productivity.

Reactor and Reactor Core Design

The SRS reactors have an extensive system of removable reactor poison that can be withdrawn as Xe-135 grows in from the decay of I-135. Each of the 61 control positions has a complement of 7 rods, 2 of which are designated for half-length rods for control of the axial power distribution. The other five are full-length rods and are withdrawn sequentially. The control rod complement is adequate to ensure that the reactor is sufficiently subcritical in its shutdown state. The full-length rods are withdrawn to bring the reactor critical, further withdrawn to compensate for the negative effect of temperature as the reactor power is increased, and further withdrawn for the negative effect of the Xe-135 poison that grows in.

The function of the SRS reactors is to produce materials, primarily tritium and plutonium, by transmutation of fertile material by neutron capture. So the reactor core is a mixture of fissionable material: namely U-235 to produce the neutrons; and of fertile material, namely Li-6 if the core is to produce tritium; and U-238 if the core is to produce plutonium. The mixture of fissionable and fertile materials must be nearly balanced if the reactor is to be able to operate, but an excess of fissionable material is necessary to allow for the effects of temperature and Xe-135. The control system compensates for the fissionable material excess.

Reactor Operation

After the reactor has operated for a few days, the I-135 and Xe-135 concentrations achieve an equilibrium level, and the poisoning effect of xenon is constant. However, if the power level is changed, the equilibrium is disturbed, and a transient in the Xe-135 concentration is generated. If the reactor is shut down, power is reduced to zero, the xenon transient is very large and in some cases cannot be compensated by the control system.

Power Level Changes

Initial Startup of a Reactor Core

Usually a reactor after startup is taken to full power in a series of steps, for example, 40%, 60%, 80%, 90%, and 100%. At each step, checks are made to determine that operations are normal. Power distribution shaping is also done with the control system.

Control rods are withdrawn to bring the reactor critical. Thereafter, the reactor remains very near critical, and control rods are moved only to compensate for other effects, such as power level changes, Xe-135 build-in, or fuel burnup. To take the reactor to 40% power, control rods are withdrawn to make the reactor slightly supercritical. The reactor responds by increasing power. The reactivity change from the associated change in temperature occurs within seconds. The 40% power corresponds to about 800 megawatts. The rate of rise is procedurally limited, so the total change takes maybe ten minutes. When the 40% level is reached, the control rod position is in equilibrium with the reactor temperature.

At the 40% level, Xe-135 starts to build in, and control rods must be withdrawn to compensate for the decrease in reactivity. The change is slow, so control rod motion can easily follow. Figure 1A is a graph of the reactivity change from Xe-135 in units of %k as a function of time. If no further change in power were made, the Xe-135 poison would reach its equilibrium level of 2 %k after about 36 hours.

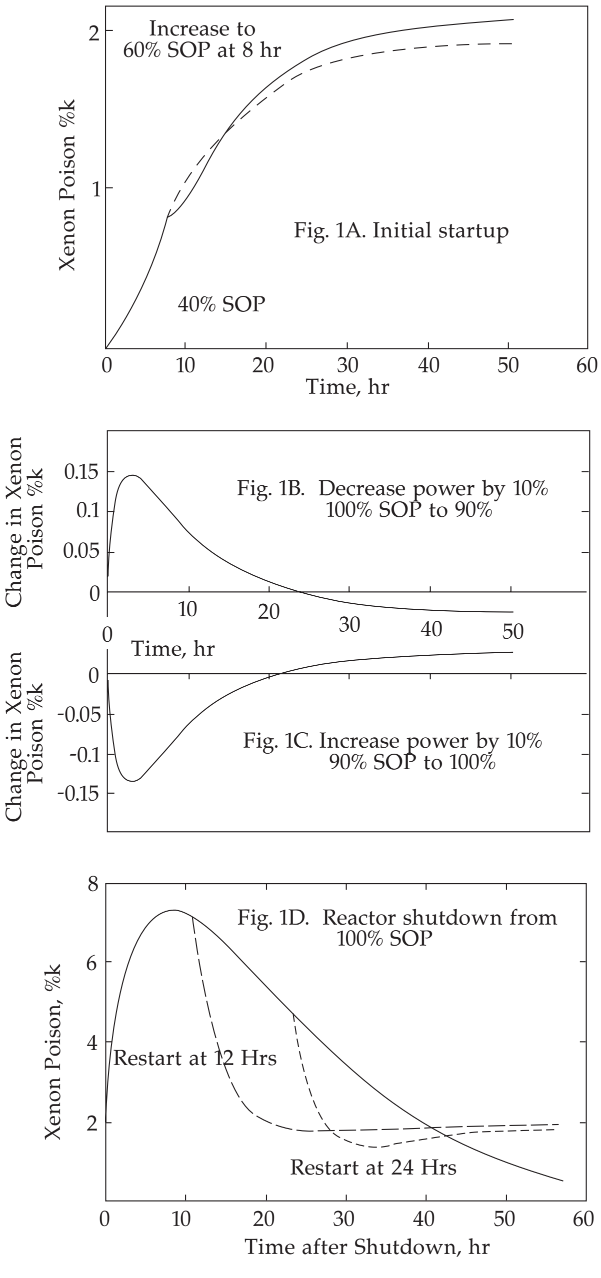

Figure 1. Xenon transients

Suppose that after eight hours the power level is raised further to 60% of full power. This is also illustrated in Figure 1A. After the rod position has equilibrated with the temperature changes, the control rod withdrawal rate is a little slower than it would have been without the power increase. The Xe-135 that had built in is burned up faster at the higher power than additional Xe-135 can grow in from the higher rate of I-135 production. But after another eight hours, the rate catches up, and the Xe-135 poison goes on to equilibrate at a slightly higher level than it would have at 40% power.

Power Level Adjustment

Suppose the reactor is at equilibrium at 100% power and for some reason it is desired to decrease the power say to 90%. This change produces a Xe-135 transient because at 90% standard operating power (SOP), Xe-135 is not burned up as fast as it grows in from the I-135 present. This transient is illustrated in Figure 1B. The increase in Xe-135 poison reaches a maximum in about three hours after the change and then gradually subsides to the new, slightly lower equilibrium level. To achieve the power level decrease, control rods are inserted over a few minutes, then withdrawn for three hours as the Xe-135 poison builds in, and then inserted as the Xe-135 transient dissipates.

If the reactor is at equilibrium at 90% power and then the power is increased to 100%, the Xe-135 transient is the reverse—nearly a mirror image across the time axis as illustrated in Figure 1C. Control rods are withdrawn over a few minutes for the power increase, inserted for a few hours for the decrease in Xe-135 poison, and then withdrawn over a couple of days to the new equilibrium level.

Reactor Shutdown and Restart

If a reactor is shut down after operating for a while at a high power level, the Xe-135 transient is large. The I-135 level is high because of the high power, and then after shutdown the I-135 decays to Xe-135 that is not burned up. The Xe-135 transient after shutdown is illustrated in

Figure 1D. The peak xenon poison occurs after about 10 hours. During the period of peak xenon, the reactor is very subcritical. Some of the reactor charges utilized at Savannah River, because of insufficient available reactivity, were limited as to the time at which they could be restarted. Others, however, could be restarted at any time.

Recovery situations are also plotted in Figure 1D. Power level at recovery is 40%. The high Xe-135 level burns up rapidly, then proceeds to the equilibrium level. For some SRS reactor charges, the recovery level was limited to 20% so that the reactivity from Xe-135 burnup is more easily managed.

Xenon Oscillations

Xenon oscillation is the name given to the redistribution of the power in local regions of the reactor caused by redistribution of the Xe-135 poison. For example, suppose the reactor is in an equilibrium state at a power level where Xe-135 poison is significant. At some time, the half-length control rods are moved down a short distance; the power in the top part of the reactor goes up and that in the bottom part of the reactor goes down. At the top, Xe-135 burns up faster than it is replaced by I-135 decay, increasing the reactivity and therefore the power in the top. After a while, the generation of I-135 at the top catches up, and Xe-135 concentration levels off and starts to increase and thereby to decrease the power at the top. The decrease in power continues until the generation of I-135 no longer replaces the I-135 that decays to Xe-135, and the Xe-135 concentration levels off and starts to decrease, and the power starts to increase. The oscillation has gone full cycle. Conditions in the reactor bottom are just the reverse.

Just described is an axial oscillation, but oscillations can occur on opposite sides of a reactor (an azimuthal oscillation) or, more complicated, a top-side vs. a bottom-opposite-side (a quadrupole oscillation). All types have occurred in the SRS reactors; examples of the experience will be discussed later. In the example of the previous paragraph, half-rod motion caused the perturbation in power distribution to initiate the oscillation. In effect, any perturbation in the power distribution will cause a xenon oscillation; its size and longevity depends on characteristics of the reactor charge and control operations.

The oscillations can be damped, in which case each succeeding cycle diminishes, or undamped, in which case the amplitude of each succeeding cycle increases. The dividing line or threshold (in terms of power density) for undamped oscillations depends on several factors that are characteristics of a particular reactor charge. Characteristics that tend to lower the threshold are: lower fuel loading (e. g., end of fuel cycle vs. beginning); greater flatness of the power distribution (e. g., greater number of fuel assemblies operating at the same power); and smaller negative temperature coefficients of reactivity. Because of the large size, high power density, and great degree of power distribution flatness, most reactor charges that have been operated at SRS are susceptible to xenon oscillations despite their negative temperature coefficients of reactivity.

Reactor Instrumentation

To adequately control xenon oscillations, it is necessary to know the three-dimensional power distribution. The reactor cooling system is comprised of six loops, each with two heat exchangers. The six loops feed a common plenum at the reactor top that supplies D_2O coolant to all fuel assemblies. The flow goes down each fuel assembly and then out the bottom into the bulk moderator space. Each of the six loops then draws flow from the bulk moderator via six exit nozzles evenly spaced around the bottom of the reactor vessel.

The primary detector for measuring the radial power distribution is the temperature monitor. There are four thermocouples associated with each fuel position. These thermocouples monitor the effluent temperature from the fuel assembly. The effluent temperature along with the plenum inlet temperature and the assembly coolant flow provides the assembly power. The on-line computer processes the data and provides a complete radial power map.

Operating off the same thermocouples is the radial power monitor. The on-line computer processes the data also to provide the average power of the six fuel assemblies surrounding each control position.

The third radial system and the best for monitoring azimuthal xenon effects on the radial power distribution is that of measuring the so-called pump suction temperatures. Thermocouples are located in the lines from the reactor effluent nozzles to the coolant pumps. The pump suction temperature along with the plenum inlet temperature and the loop coolant flow provides a measure of the power in that one-sixth sector of the reactor.

The instrumentation for determining the axial distribution is a system of nine rods, each placed interstitially among three fuel assemblies. The nine rods are located strategically throughout the reactor so as to provide complete coverage. Each rod contains seven gamma thermometers and a thimble for irradiating a wire. The gamma thermometer is a small iron pin attached to a heat sink at one end. Thermocouples are attached to each end of the pin to measure the temperature difference between the ends. This temperature difference is proportional to the gamma ray energy flux at the location of the pin and therefore to the fission rate and power generation in the vicinity of the pin. The seven gamma thermometers in a rod provide a good measure of the axial power distribution. The wire irradiation with subsequent measurement of the gamma ray activity along its length measures the axial neutron distribution and is used to periodically calibrate the gamma thermometers.

Operating Experience

Under normal conditions, xenon effects are not specifically monitored. Close control of the power distribution is maintained as part of standard operations and is usually sufficient to eliminate problems from xenon. Circumstances under which xenon problems are encountered occur infrequently and do so in charges that are unusually susceptible to oscillations with damping factors less than unity. Examples of xenon oscillations are given below.

The clearest example of an inadvertent undamped oscillation is illustrated in Figure 2A. This occurred during power ascension when control of the power distribution is relaxed somewhat from the requirements at full power. The ordinate is the change in one pump suction temperature from an equilibrium value in arbitrary units; this was an azimuthal oscillation so that the oscillation on the opposite side of the reactor was 180 degrees out of phase with the illustrated oscillation. The arrows indicate points at which control rods were moved; the motion was not sufficient to control the oscillation. Control at the end of about 50 hours required a relatively large amount of control rod trim.

This illustrates that, even though there is sufficient time for control, sufficient and correct control procedures must be supplied. The method of control in this case was to force the perturbation to zero and hold it there. This works, but requires a larger amount of reactivity change than the method illustrated in the next example.

The most complex and difficult-to-control oscillation to occur had a quadrupole nature. This oscillation was in the axial direction, but the oscillation on one side of the reactor was 180 degrees out of phase with the oscillation on the other side of the reactor. Figure 2B shows the oscillation in terms of the change in relative

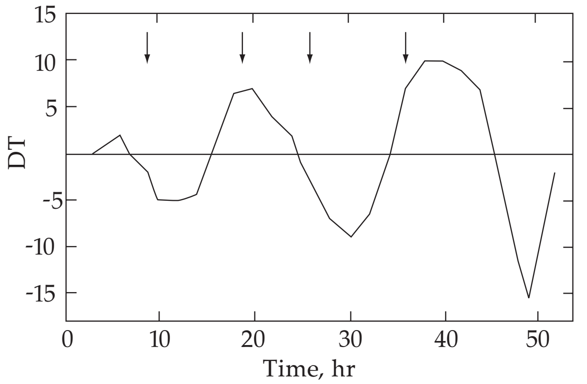

Figure 2A. Observed undamped azimuthal oscillation

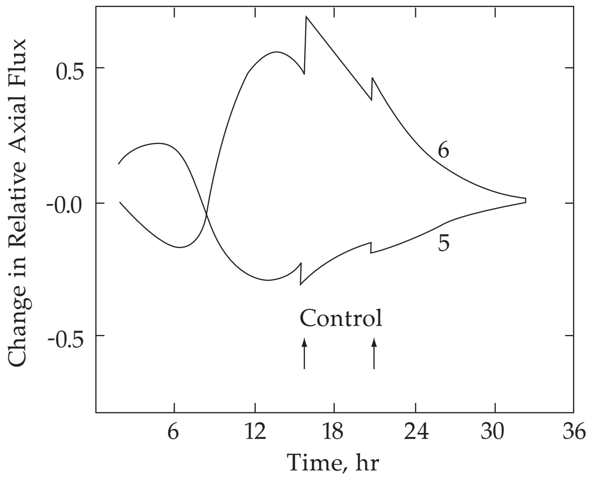

Figure 2B. Quadrupole oscillation

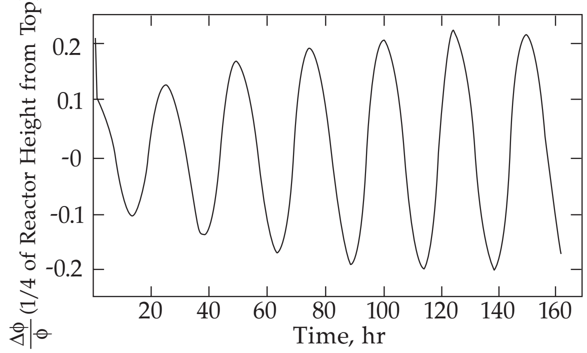

Figure 2C. Axial xenon test in SRS reactor

Figure 2. Xenon oscillations

axial power in the top of the reactor on the two sides as measured by the axial power monitor (gamma thermometer rods); the numbers at the two curves are the numbers of the indicating gamma thermometer rods. To damp the oscillation, partial rods in five septifoils in the vicinity of #6 were moved down. At the same time, partial rods in five septifoils in the vicinity of #5 were moved up. This illustrates a more efficient method of bringing under control an oscillation that has become large. The coming action is anticipated and counterbalanced. In regions where the power is high but is starting to decrease, indicating that xenon is growing faster than it is burning up, rods are moved to keep the power high in order to burn up xenon. A relatively small amount of reactivity is required.

A series of controlled experiments with deliberately induced xenon oscillations was carried out in one of the SRS reactors. The majority of the tests dealt with axial oscillations and a few with azimuthal oscillations. The results of one of the more interesting axial tests is shown in Figure 2C. The ordinate is the change in power at a point one-quarter of the way from the top of the reactor under conditions of constant reactor power. The oscillation diverged for a few days and then appeared to saturate for an unknown reason.

The phenomenon of xenon oscillations is an interesting one in that the theoretical description is simple and remarkably accurate. The theoretical aspects are not treated here, but extensive theoretical studies were made and were a great assistance in dealing with the phenomenon.

Effect of Xe-135 on Productivity

The purpose of the Savannah River Site reactors is to produce materials not found in nature in recoverable quantities. They do this by utilizing neutrons produced from fission of U-235. At the high power density levels at which the SRS reactors operate, only a small fraction of the Xe-135 decays radioactively; most is destroyed by neutron capture. These neutrons otherwise could have been used in production processes.

Consider the production of tritium, an isotope of hydrogen with an atomic mass of three.

The production process is

Li-6 + neutron —> H-3 + He-4.

A neutron is absorbed by an atom of Li-6 and the resulting Li-7 splits into a tritium nucleus and a He-4 nucleus plus energy. The energy is carried off by the two resulting particles and dissipated in the substrate material.

Reactor charges for production of tritium are composed of uranium highly enriched in U-235, lithium enriched in L1-6 contained in suitable fuel, target elements, and assemblies. The elements and assemblies use aluminum as the diluent and encasing material.

For greatest efficiency, it is desirable to maximize the number of neutrons available to the production process. Impact of Xe-135 can be understood from the "neutron economy".

Fission of an atom of U-235 produces on the average 2.43 neutrons. Uranium-235 also absorbs neutrons to produce U-236 so that each neutron absorbed by U-235 produces 2.08 neutrons after accounting for the U-236. Since one neutron must be available for absorption in U-235 for the next generation, 1.08 neutrons are potentially available for the production process.

Other processes subtract from the neutrons available for production. For the SRS reactors, about 3% of neutrons leak from the reactor core and are absorbed by the reactor tank wall or the reactor shields. This takes 0.06 of the neutrons from the neutron economy. (Use of only lithium-bearing assemblies in the outer ring of the reactor and having the target elements extend above and below the fuel elements utilize some of the leakage neutrons for production.)

The neutron moderator, D_2O, the aluminum in the fuel, and target assemblies absorb another 5% of the available neutrons. This subtracts 0.10 of the neutrons from the neutron economy.

During the lifetime of the reactor core, fission products build up in the fuel assemblies. The fission products (including Sm-149) other than Xe-135 absorb about 3% of the available neutrons; this subtracts another 0.06 from the neutron economy.

Xenon-135 is essentially all destroyed by neutron capture. Since each fission produces 0.063 atom of Xe-135, the Xe-135 subtracts 0.06 from the neutron economy.

The remainder 2.08 - 1.00 - 0.06 - 0.10 - 0.06 - 0.06 = 0.80 neutron per fission is available for production of tritium. The neutron economy may be summarized as shown in Table 2.

Table 2. Neutron Economy

	Production	**Utilization**
Fission	2.08	
Next generation		1.00
Leakage		0.06
Absorption in D_2O and structure		0.10
Fission products other than Xe-135		0.06
Xe-135		0.06
Production		0.80
Total	2.08	2.08

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Biography

With a Ph. D. degree in Experimental Physics from Indiana University, Mr. Roggenkamp joined the Du Pont Co. on October 1, 1952, in the reactor training program at the Argonne National Laboratory. Coming to Savannah River the next year, he joined the Experimental Physics Division of the Savannah River Laboratory (SRL). During the period of 1957 to 1964 in the Reactor Technology Section of the Savannah River Plant (SRP), he supervised work on the physics aspects of the nuclear reactor operations. During this period of time, production of Pu-238 was begun. Returning to SRL in 1964 as Research Manager of the Theoretical Physic Division, he was instrumental in the development of the high flux demonstration program, and of the curium and californium production programs. During the ten years prior to retirement in December 1987, he was in charge of the Advanced Operational Planning Division of SRL, which provided long-range studies for operation of SRP and for the integration of SRP operations into the Department of Energy complex.

The Nuclear Test Gauge

Thomas F. Parkinson and Norman P. Baumann

Abstract

Safe and efficient operation of the Savannah River Plant (SRP) production reactors required strict quality control of all fuels and targets that were loaded into the reactors. To analyze the composition of these components in a "nuclear environment," a large low-power critical facility, the 305 Test Pile, was built. This facility required about 10 minutes per test and also required a relatively large, well-trained crew operating under strict and extensive procedures.

The Nuclear Test Gauge (NTG), a small, slightly subcritical facility, was developed to analyze production reactor components about ten times faster than the 305 Test Pile with comparable accuracy and with a much smaller operating staff. After about 20 years of successful experience with the NTG, an extensive modification program was undertaken, which resulted in major improvements. The resulting "mini-NTGs" accelerated component testing with improved safety margins by operating much further below criticality. The mini-NTGs also incorporated improved instrumentation and neutron sources.

Introduction

The design and operation of the five Savannah River Plant (SRP) production reactors demonstrated an extraordinary capability to escalate power and to produce a wide variety of isotopes. Achieving this capability required developing a variety of fuel and target designs that facilitated an almost eightfold increase in reactor power. A vital factor to ensuring safe reactor operation was an efficient and accurate quality control methodology for all the fuels and targets that were charged into the reactors.

Later, the Nuclear Test Gauge (NTG) was developed at the Savannah River Laboratory (now the Savannah River Technology Center) to supplement and eventually replace the 305 Test Pile. U.S. Patent No. 2,936,274 was awarded to Gerhardt Dessauer for this concept.

When the Hanford reactors were built during World War II, nuclear testing of reactor loadings was accomplished by measuring the reactivity effect of each component using a large, low-power critical assembly—the 305 Test Pile (Cawley 1955). This facility was graphite-moderated and fueled with natural uranium slugs. It required a highly trained crew operating under extensive procedures. Each component test required about 10 minutes. When SRP was constructed some 10 years later, an identical 305 Test Pile was built and operated in the SRP fuel and target manufacturing area.

Preliminary Experiments

A small research reactor, the Standard Pile (SP) located in Building 777-M, was used to investigate the feasibility of a subcritical test facility. The SP was designed and constructed by the General Electric Company and was similar to the Thermal Test Reactor at Knolls Atomic Power Laboratory (Stewart 1953). Both reactors were graphite-moderated and used aluminum alloy fuel containing uranium highly enriched in the U-235 isotope. The critical mass was only 2.3 kg of U-235, making the SP highly sensitive to small variations in test pieces inserted into the center of the reactor core.

The objective of the preliminary experiments was to establish a subcritical operating regime wherein the response time to small changes in the composition of test samples was rapid while the resultant changes in the neutron population were accurately measurable. As a subcritical reactor containing a neutron source is brought closer to criticality, the response time increases from the effect of the delayed neutrons within the fission process. Simultaneously, the neutron multiplication, M, increases so that the neutron flux produced in the reactor core can be measured with increasing precision. The criticality status of a nuclear reactor is characterized by the effective multiplication factor, k_{eff}.(1)

From experiments conducted in the SP, it was concluded that k_{eff} values between about 0.98 and 0.99 (corresponding to neutron multiplications of 50 to 100, respectively) would be appropriate to achieve the required compromise between response time and sensitivity.

The next stage in the evolution of a production testing facility was to construct a working prototype. Readily available fuel, components, and instrumentation were used to expedite this development. Nuclear safety was paramount throughout the entire development program. The prototype NTG was installed in a shielded room in Building 777-M, so that any operation involving close approaches to criticality could be carried out remotely. The most-suitable fuel elements available were aluminum alloy cylindrical rods containing highly enriched U-235 and measuring about 1 inch in diameter and 12 inches long (Mark II fuel rods).

A great advantage of these fuel rods was that experimental data were available from the Oak Ridge National Laboratory on the optimum spacing for a triangular lattice array in light water (H_2O) moderator. Thus, if any distortion of the core occurred (e.g., from the impact of a falling object), k_{eff} would decrease.

Figure 1 shows the core assembly of the prototype NTG. The core components were mounted in a 55-gallon drum, and shielding was provided by a thin cadmium sheet that absorbs thermal neutrons and by concrete blocks to absorb gamma radiation. A through tube was located in the center so that test specimens of known composition could be inserted. A Po-Be neutron source emitting about 10 million neutrons per second was mounted near the core center. Two boron-lined ion chambers were suspended in the fuel drum; one chamber actuated the safety system and one chamber supplied a signal used to detect small changes in the neutron flux corresponding to changes in the composition of test specimens. Three other external neutron-counting systems monitored the neutron flux leaking from the core tank.

The safety system consisted of a cadmium safety blade suspended above the core by an electromagnet, which released the blade automatically in the event of an unexpected increase in the neutron flux. The safety blade could also be released by a push-button on the instrument rack.

After construction of the prototype NTG was completed and tested, fuel was added incrementally until a neutron multiplication of about 55 was attained. About 4 kg of U-235 were required. The assembly was moderated by deionized H_2O and had H_2O reflectors about 10 cm thick on the sides but no reflectors on the top or bottom of the core.

Experiments with the prototype NTG demonstrated that a sensitivity could be obtained that was adequate for production tests of reactor components. The experience derived from constructing and operating the prototype NTG provided vital information for designing a permanent model.

Much of the sensitivity of the NTG is from its small size relative to the 305 Test Pile. Thus, the fuel content of a test sample is a much larger fraction of the NTG fuel loading compared to the 305 Test Pile. Additionally, the NTG ion

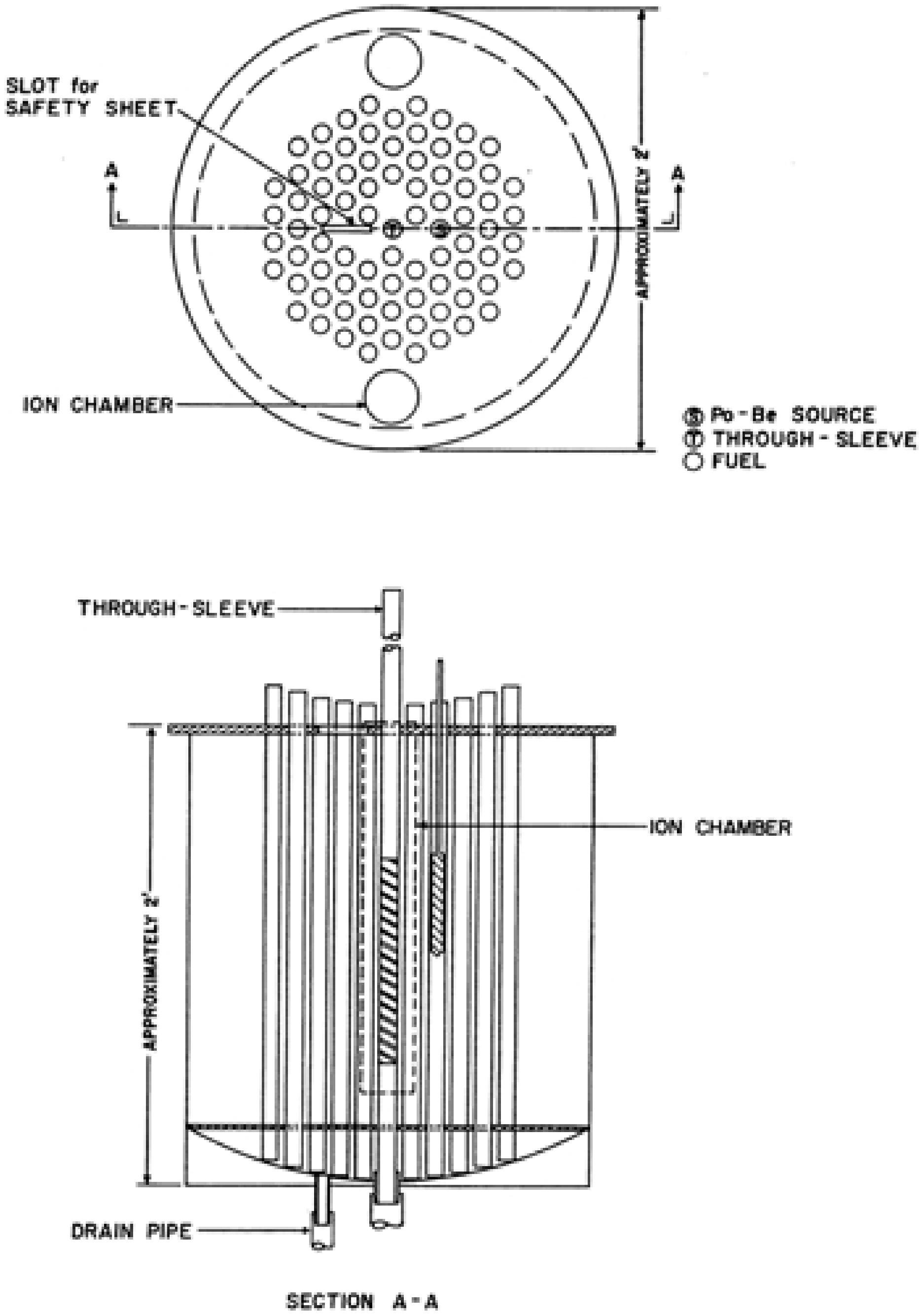

Figure 1. Prototype NTG

chamber absorbs a much larger fraction of the total neutrons produced than the 305 Test Pile ion chambers. The precision achieved in sample analyses could be enhanced by using a stronger neutron source and by efficiently collecting thermal neutrons leaking from the core.

A complete description of the prototype NTG is given in the reports by Axtmann, Dessauer, and Parkinson (1955a and 1955b).

Production Model NTG

Design Features

The basic core design of the prototype NTG was preserved in the production model except that the core axis in the latter was horizontal rather than vertical. This change was made to simplify the sample feed system for the production model.

To facilitate testing a wide variety of fuel and target designs, the production NTG core had a hexagonal polyethylene plug that could be removed from the core and replaced with plugs, which could accept more advanced fuel and targets designs.

The single cadmium safety blade was replaced with two large boral safety sheets in the production model NTG. These sheets were automatically dropped into the core in the event of an unexpected increase in neutron flux. In addition, a dump valve was provided in the production model that could automatically drain the H_2O moderator to the midplane of the lattice in five seconds.

Based on radiation surveys of the prototype NTG, the shielding of the production model was designed to provide a safe radiation environment for operations personnel. (See Figure 2.) A concrete wall 2 feet thick and 9 feet high surrounded the entire system except for open-

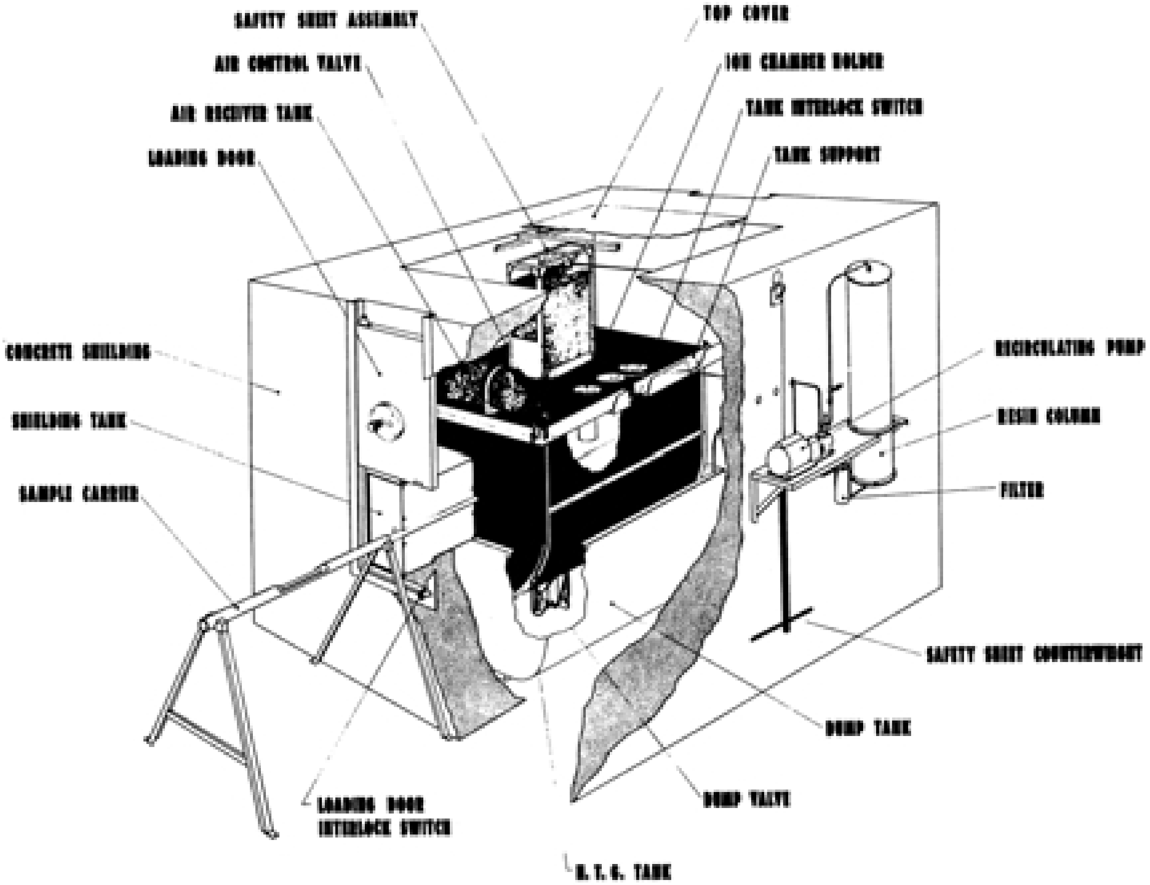

Figure 2. Production NTG

ings on the top and two sides. The inner concrete surface was lined with cadmium sheet, and access to the front and rear of the core tank was via removable H_2O-filled shield tanks and by lead-filled loading doors.

Neutron detection was achieved by six ion chambers lined with boron-10 and suspended in the H_2O-filled tank, which contained the core. Two of the ion chambers provided input to the emergency shutdown system. The other four ion chambers were connected in parallel and fed a differential electrometer capable of detecting the small changes in current corresponding to changes in the composition of test samples.

The emergency shutdown system was actuated by two independent circuits that caused the two safety sheets to fall within one-third of a second and that in turn opened the dump valve and drained the core tank. Rapid shutdown also resulted if the loading doors were raised during operation of the system.

A Po-Be neutron source was installed in one of the fuel tubes near the core center. This source initially emitted about 30 million neutrons per second. The emission rate decreased with a 138-day half-life during the course of the initial calibration experiments and subsequent production testing. Consequently, carefully pre-

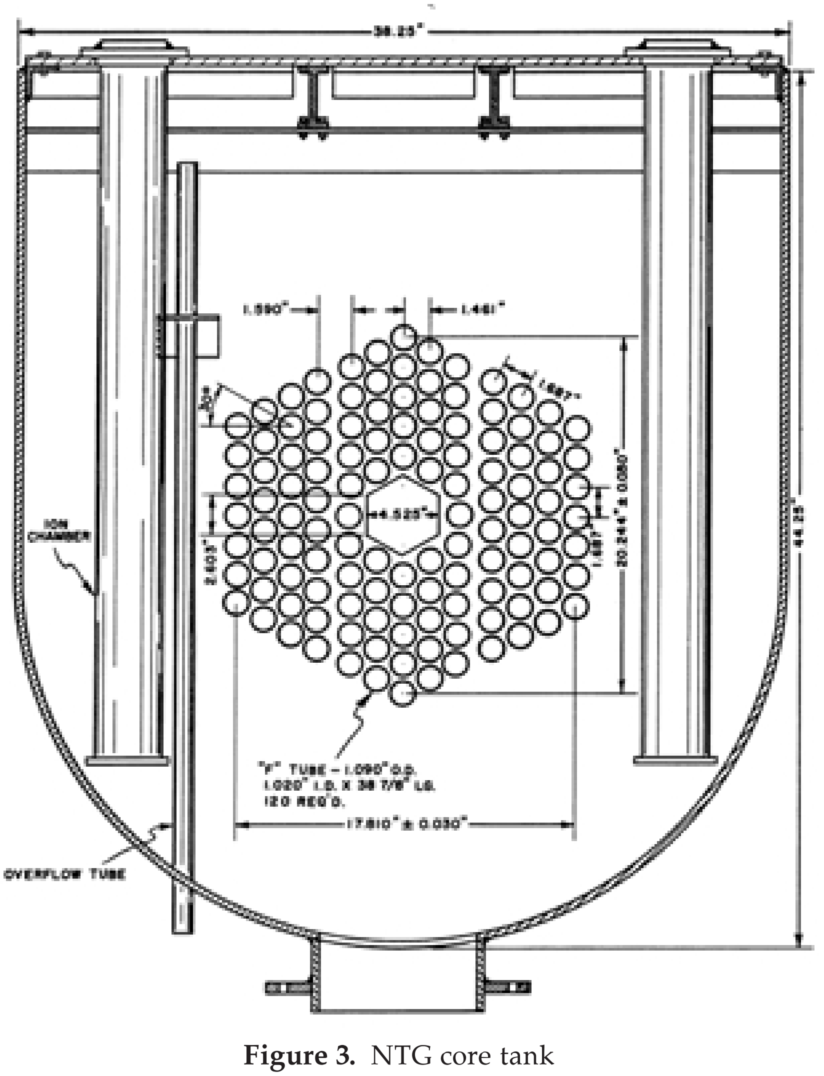

Figure 3. NTG core tank

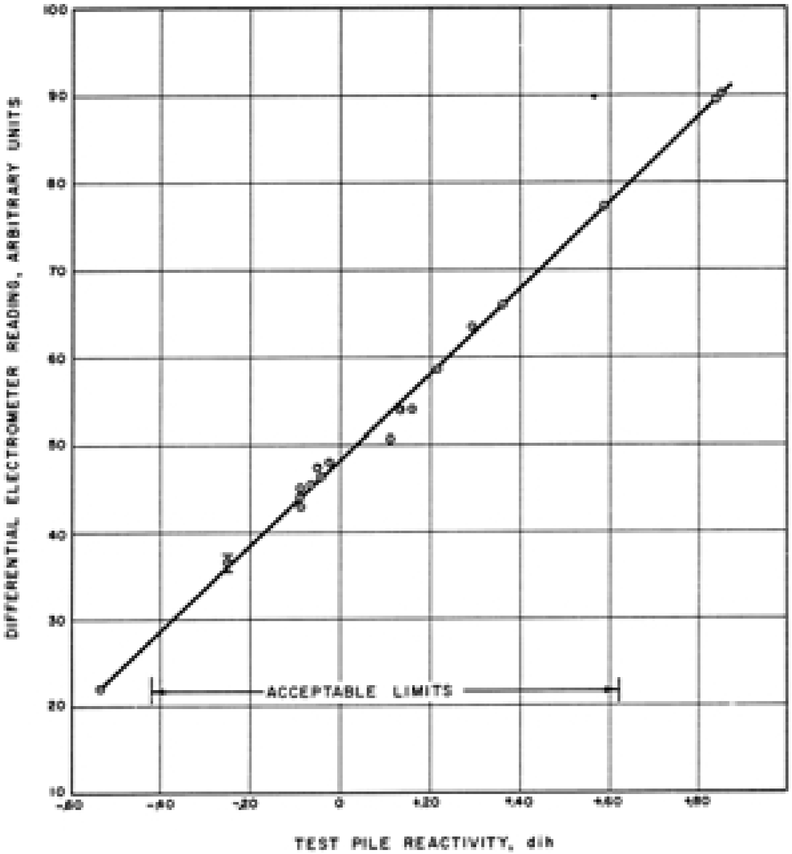

Figure 4. Correlation of NTG and 305 Test Pile data

pared standards were essential to periodically calibrate the NTG. Subsequently, a longer-lived Ra-Be neutron source was substituted for the Po-Be source.

Performance

A remarkable feature of the five SRS production reactors was their capability for power escalation. This capability required an extensive development program to manufacture advanced fuels and targets. Accordingly, the production model NTG had to have the versatility required to test components of vastly different geometries from the initial Mark I natural uranium fuel for plutonium production through the Mark 22 extruded aluminum alloy fuels and targets for tritium production. As shown in Figure 3, the removable octagonal plug provided the requisite versatility.

The NTG tests do not provide an absolute analytical method, so testing of core components depends on calibration with standards of known composition. Initial calibrations utilized existing standards prepared for the 305 Test Pile. Numerous correlations were obtained between NTG and Test Pile data to ensure that no loss in precision occurred with the former. Typical results of these correlations are shown in Figure 4. In addition, it was demonstrated that testing in the NTG was about 10 times faster than testing in the 305 Test Pile, and that the staff required for the NTG was only about 20% of that required for the Test Pile. Substantial cost savings resulted.

For the extruded fuel and target tubes, standards were fabricated from ingots whose compositions were known from material balance and whose concentrations covered the

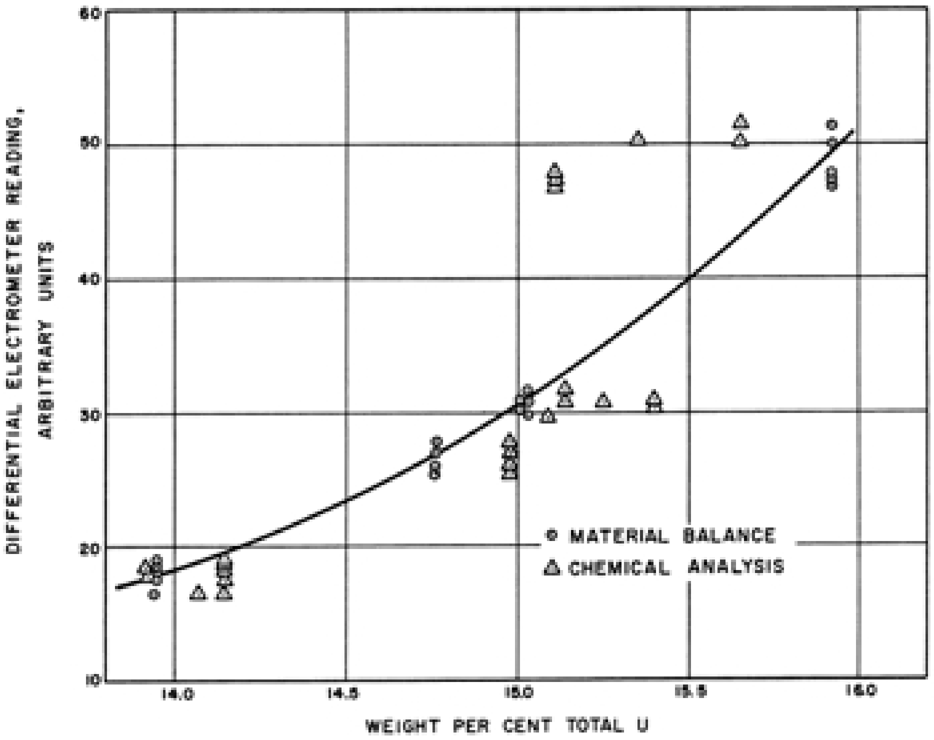

Figure 5. Calibration for tubes of enriched U-Al alloy

range to be expected from production runs. Small samples were then taken from the extruded tubes for chemical analysis. However, the variations in the chemical analyses compromised their usefulness. Thus, the material balance values represented the most reliable data for the calibration standards. A typical calibration curve is shown in Figure 5. A complete description of the NTG is given in Parkinson et al. (1956).

The New Mini-NTGs

The original design of the NTG served its function well, but over time two related major shortcomings became apparent. First, the NTG could not perform all of the functions of the 305 Test Pile, so it was necessary to continue to operate both facilities. Second, the NTG operated so close to criticality that it had to be designed and operated much as a critical facility. This included a SCRAM system, a water dump capability, and several mechanical safety devices. In addition, as critical facilities, both the NTG and the Test Pile required a large investment in highly trained manpower and time consuming procedures. After the initial installation, little was done to upgrade nuclear instrumentation or neutron detectors. Over the years, nuclear instrumentation had improved dramatically, so exploitation of these improvements was clearly indicated. In addition, mini- and micro-computers became available.

The Low-k_{eff} Concept

The original NTG had a reference multiplication constant k_{eff} of about 0.96 to 0.98. With highly enriched fuel inserted, criticality could be closely approached. Computations showed that if the reference k_{eff} could be reduced to about 0.84, no conceivable misloading and flooding could result in a k_{eff} above 0.95. Thus, the facility could be totally designed and operated without consideration for criticality safety, and no special operator training would be required. Computations supported by reactivity tests in the NTG showed that the low k_{eff} could be attained by removing either the inner or the outer ring of U-235 fuel slugs and replacing them with polyethylene rods of the same diameter. The major problem of low-k_{eff} operation was the reduction in sensitivity. This was a combined result of a lowering of the reference neutron flux and of the smaller percent neutron flux change caused by insertion of a test component. The sensitivity was not only recovered, but was enhanced by a sequence of modifications:

- The two ion chambers previously devoted to the SCRAM circuit were devoted to data acquisition.
- Replacing the Ra-Be source with its intense gamma rays with Cf-252 sources eliminated the need for compensated ion chambers. Uncompensated chambers with boron coatings on all surfaces doubled the current (and were more stable as a bonus).
- The biggest contribution was obtained by revamping the method of data taking. The original NTG assayed fuel and target tubes by moving them in incremental steps. A full-length tube was inserted, a waiting time established, and a reading taken. The tube was then advanced and the process repeated. Only a fraction of the time was spent recording usable data. Most of the time was spent waiting for the delayed neutrons to come to equilibrium. Low-k_{eff} operation greatly reduced the time and relative magnitude of the operation. It was possible to eliminate the incrementing, replace it with a continuous uniform speed drive, and thus use all of the ion chamber current.
- For small samples, with a small effect on reactivity, the desired sensitivity was obtained by using the “pile-oscillator” technique. In this procedure, the test sample is repetitively inserted and withdrawn over an extended time period to obtain both statistical accuracy and compensation for slow drifts in the response of the system. This system was used for “bottle samples” containing solutions of gadolinium and boron as well as heavy water samples. These assays were eventually relegated to a neutron blackness tester.

Analysis

The original analysis method consisted of reading off values from a calibration curve drawn manually through calibration points plotted for grams per foot values assigned to the standards. This procedure was replaced with the “NTG Index” that was simply the fractional change in neutron flux level caused by introducing the sample relative to the reference flux with the test port empty. This value is independent of the source strength and small drifts in reactivity and instrument sensitivity. Moreover, it can be verified at a later time if necessary.

This Index has a simple relation to the effective multiplication constant, k_{eff}, for the system. The value of k_{eff} can be calculated from various reactor codes (e.g., ANISN). In general, the ANISN calculations do not exactly agree with the indices measured for the standard, but if the k_{eff} values are normalized to give the best fit to the standards over the range of interest, a good fit to the standards is obtained with a shape much more accurate than simply fitting to the measured points. Finally, the ANISN values are fitted to a low order polynomial for g/ft as a function of the Index.

Implementation

The physics design of the new NTG closely matched the original with respect to dimensions, lattice pitch, and H/U-235 ratio. The water tank was replaced with a cubical block of water extended polyester (WEP). The test port and cylindrical channels for the fuel were drilled into the WEP block. Water retention was obtained by coating all surfaces with a lacquer impervious to water vapor. Separate NTGs were built for Building 321-M (enriched fuel) and Building 320-M (targets). Locating a separate NTG in Building 321-M has obvious safeguards advantages, but separate units also enabled different optimum core designs to be used for each. For Building 321-M, the outer ring of fuel was replaced by 1-inch-diameter polyethylene rods; for Building 320-M, the inner fuel ring was removed and replaced instead. The latter configuration had the advantage of greatly increasing the sensitivity to U-235 and greatly reducing the sensitivity to neutron-absorbing atoms such as U-238, U-236, and U-234. A result was that small calculated corrections could be made for variations in isotopic content rather than requiring standards with varied isotopic content.

Standards

With the improvement of the sensitivity (i.e., precision), it became apparent that uncertainties were due primarily to the accuracy to which the content of the standards was known. Various methods, both destructive and non-destructive, were explored to develop new methods for assay of the standards. Of these, a non-destructive assay based on thermal neutron transmission was the most successful. An assay could be made using the measured transmission along with known thermal neutron cross sections and a geometrical description of the tube.

Conclusions

For over two decades, the Nuclear Test Gauge served a vital function in the quality control of many thousands of components that were irradiated in the five SRS production reactors. The success of the NTG resulted in cost savings of millions of dollars. The NTG served SRS well from its initial installation until the reactors were shut down. The NTG was essential for the implementation of co-extruded tubes (i.e., Mark-16) in reactor charges. The NTG provided the data for estimating initial critical configurations as well as providing the data for proper internal heat splits in the fuel and target matching computer programs.

Starting in 1978, an extensive effort was made to improve the safety and performance of the NTG. To avoid any credible accidents that might cause a super-critical excursion, some fuel rods were removed from the NTG cone. Improved instrumentation and improved neutron sources enabled faster and more accurate analyses to be achieved.

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End Note

1. The status of a nuclear reactor is characterized by k_{eff}, the effective multiplication constant. If k_{eff} is less than one, the reactor is subcritical, and the neutron population falls to zero unless a supplementary neutron source is

provided. If k_{eff} is exactly one, then the reactor is critical, and the neutron population is constant since the neutrons produced by fission are just equal to the neutrons lost by absorption and leakage from the reactor. If k_{eff} is greater than one, the reactor is supercritical, and the neutron population increases.

In a subcritical reactor like the NTG, a supplementary neutron source is provided, and this neutron source is multiplied by a factor, M, given by

$$M = \frac{1}{1 - k_{eff}}$$

Biographies

Thomas F. Parkinson

After completing his undergraduate degree at Auburn University, Mr. Parkinson worked for nearly two years at Oak Ridge before enrolling at the University of Virginia. He completed his Ph.D. in Physics in late 1952 and started work for DuPont at the Savannah River Laboratory (SRL) in experimental reactor physics and instrument development. In 1960, he joined the faculty of The University of Florida, College of Engineering. Mr. Parkinson was a Fulbright Fellow in Madrid during 1966-67, after which he was appointed Nuclear Engineering Chairman at the University of Missouri-Columbia. In 1975, he joined the faculty at Virginia Tech, and after his retirement in 1990, he was designated Professor Emeritus. Mr. Parkinson is a Fellow of the American Nuclear Society and served as the first president of the Alpha Nu Sigma honor society. He made numerous visits to Argentina as a technical expert for the International Atomic Energy Agency.

Norman P. Baumann

Mr. Baumann received a Ph.D. in Nuclear Physics from Kansas University in 1954. He worked in experimental reactor physics and nuclear instrumentation from 1954 to 1993 at the Savannah River Laboratory. His expertise is in lattice physics, criticality control, reactor dynamics, activation analysis, and non-destructive analysis, including technical support of NTG operations. Mr. Baumann retired in 1993 as senior advisory scientist at SRS.

Reactor On-Line Computer Applications

Kris L. Gimmy

Abstract

Thirty-five years ago, most people were just becoming aware of the digital computer, in the form of large IBM machines. But, there was another kind of computer called the "on-line computer" just starting to be used for industrial automation. These computers looked promising as a way to improve productivity and safety at the Savannah River Site (SRS). Four technical support groups worked together to apply on-line computers to the operation of SRS reactors. The reactors were chosen because each reactor had over 3500 instrument signals that had to be monitored for proper operation.

The 20-year "computerization" effort went hand-in-glove with the effort to raise reactor powers and to produce a variety of different products. The first ten years saw computer application move from data processing, to monitoring the hydraulic limits on each reactor element, to computer control of reactor power operation. The second ten years saw newer, faster computers used as the primary safety system for reactor emergency shutdown. On-line computers were also used to automatically diagnose plant alarms and to display corrective action to the reactor operator.

Computerization was an integral part of safe operation as the reactors were upgraded to operate at seven times their original output!

Introduction

On-line computers are rugged industrial computers that are wired in to a plant process. They are there to make something run better or safer. On-line computers accomplish this by reading signals from plant instruments and running software that can operate plant controls and alarm panels. They can also do data processing and display the results to the plant operators. The on-line computers are not, however, general purpose computers, like the PC on your desk. The user cannot load new programs or alter the software designed for the specific installation (see Figure 1).

A good example of an on-line computer is the "on-board" computer in your new car. It reads about 30 signals from the engine and the environment. It adjusts for existing conditions, such as temperature and altitude, so there is no stumbling or hesitation upon startup. As you drive, it continually fine-tunes the engine, which now gives twice the gas mileage of 1950s' cars. The onboard computer improves safety in emergency conditions by controlling the brakes to avoid skidding (anti-lock brakes). It alerts you to problems with dashboard alarms. Finally, if the car needs service, it saves data that helps the technician diagnose the problem.

In like manner, the productivity and safety of the nuclear reactors at the Savannah River Site (SRS) were improved by the addition of on-line computers in an aggressive program that started 35 years ago!

1964—Data Processing and Alarms for Plant Operators

The first application of an on-line computer for SRS reactors was to do data processing and to alert operators to bad instrument signals and other minor process problems. While this sounds like a straightforward computer task, there were three major hurdles to overcome.

- A production reactor had over 3000 instrument signals that the computer would have to read.

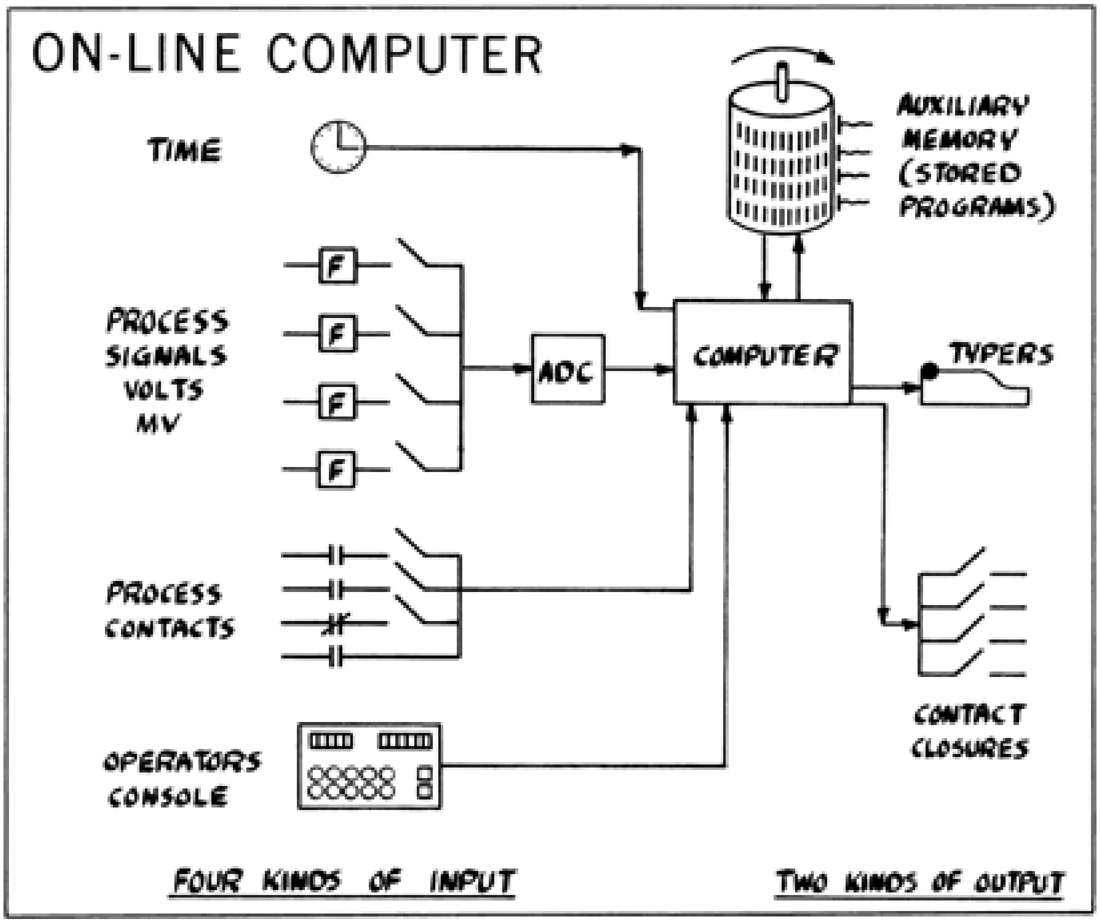


Figure 1. On-line computer

- An operator interface would have to be developed that the reactor operators could use and that would be consistent with plant procedures.
- The on-line computer system would have to be reliable and repairable by plant instrument people.

These challenges were met by four SRS departments closely working together.

The Equipment Engineering Group developed a switching matrix that would preselect reactor signals in groups of 30, which the computer could then read at 10 inputs per second. This yielded a 5-minute scan of the entire reactor process—a major improvement over the 2-4 hours it took to manually read data from recorders and the plug-in jack panel.

The Plant Project Group designed the installation of the computer (five cabinets each the size of a refrigerator) and the wiring requirements and blueprints to connect the thousands of plant signals.

The Plant Instrument Group did the wiring during scheduled reactor shutdowns without disrupting the existing analog instruments. This group also set up training and spare parts so they could repair the computers.

The Reactor Technology Group worked with the vendor (General Electric) to design a simple panel that the operators could use. Remember, this was 20 years before the Macintosh computer introduced the graphical interface we all use today. The panel specified enabled the operator to call for programs, or enter data, by using rotary switches (0-9). Two printers were also installed in the control room, one for requested data and one dedicated to alarm messages.

With the design firm and installation underway, the Reactor Technology Group took training to prepare the reactor software in the Assembler Language used by the GE computers. By the time the installations were completed, there were about 10 programs to print reactor data and display the power distribution within the reactor. The favorite program of the reactor operators was the "Histogram" program used to fine-tune the temperatures of the fuel elements surrounding the 61 control rod groups in the core. This program sorted the 1464 temperatures and generated a bar chart that showed which control clusters were hot and which were cool. It had been an onerous job that took the operators eight hours to do at the jack-panel with paper and colored pencils. The on-line computer did the job in five minutes, if you set the "Program" knobs to "08". By the end of 1964, the prototype installation at K Reactor was scanning more signals than any computer in America (including those at NASA).

1968—Automatic Monitoring of Hydraulic Limits

From day one, the SRS reactors were operated under strict, written limits to ensure there would be time for the automatic shutdown mechanisms to work if there were a process upset or equipment failure. This safety margin was monitored manually by plugging into the signals for the hottest fuel assemblies and watching the continuous traces. But, the simple temperature limits of early operation became more complex as engineering changes were made to each reactor to increase productivity. One of the major justifications for the on-line computers was to provide automatic calculation and monitoring of the hydraulic limits specified for each reactor load. Reliable monitoring of the hydraulic limits had to go hand in hand with engineering changes that eventually led to a sevenfold increase in reactor productivity. (See "Reactor Program for Increased Production Capability" by James M. Morrison in this proceeding.)

One of the most complex limits, needed at high power densities, was to provide a Burnout Safety Factor. You can visualize this phenomenon of heat flux burnout by imagining an aluminum pan filled with water, sitting on a burner on your stove. As you turn up the heat, small bubbles of steam form on the bottom of the pan, then rise toward the surface. If you turn up the heat more, the bubbles get bigger. If the heat on the pan is increased enough, the bubbles will join and form a blanket of steam on the bottom of the pan. The bottom of the pan will melt, even though the pan is still full of water.

The Burnout Safety Factor guarded against a power density high enough to cause film boiling on the aluminum cladding used on SRP fuel assemblies. Monitoring the safety factor was a complex calculation that determined the power profile along the length of each reactor fuel assembly. The formulas used data on temperature, flow, pressure, and in-core flux profiles. When done by hand, the calculation took a desk calculator and a worksheet that looked like an IRS tax form (see Figure 2). But, by 1968 (just as power was increased enough to need this limit), it was being calculated automatically by the on-line computer.

The Limits program was set to run automatically every five minutes and could not be turned off. If any parameter exceeded its limit, there was an alarm message issued. If the limit was exceeded by more than one degree, the computer closed a relay to cause a power setback of about 2%. Every hour, a summary of the margin from all of the hydraulic limits was printed automatically as a record of reactor operation.

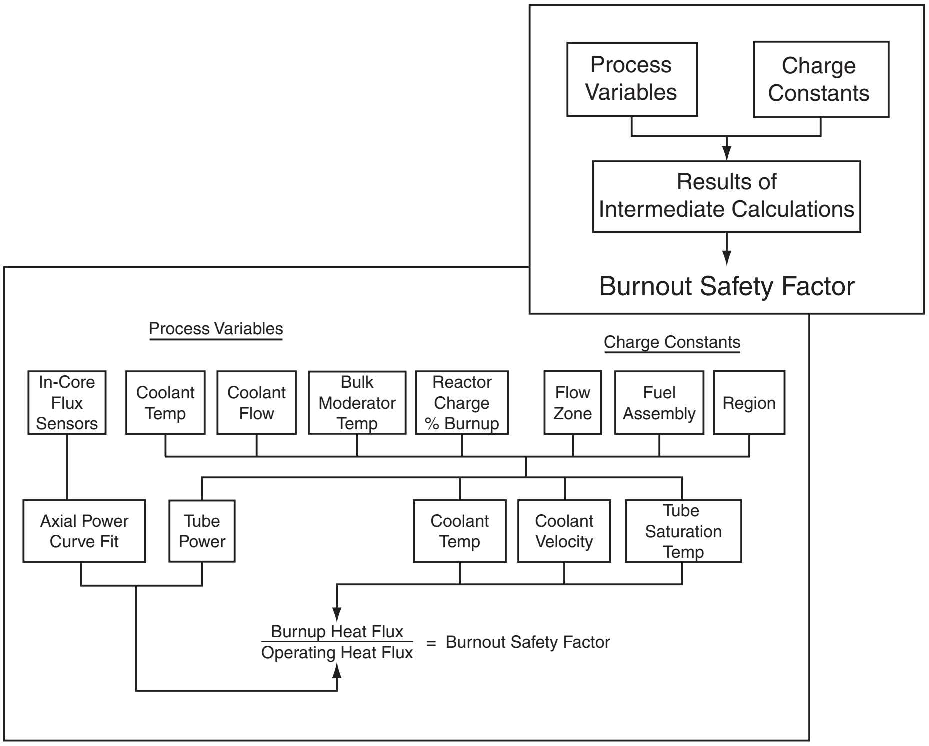


Figure 2. Burnout Safety Factor calculation

1970—Closed-Loop Control of Reactor Power

Closed-loop control of reactor operation was accomplished by adding stepping motors to the six control units used by the operators to move the three gangs of full-length control rods and the three gangs of partial-length control rods. The full rods controlled the power across the radius of the reactor, and the partial rods were shuttled up and down to control the axial distribution of reactor power.

Once again the SRS Equipment Engineering Department designed an electronic interface so a computer command could move control rods a specified number of steps. They also provided a “control panel” so the operator could select three states of automatic control (see Figure 3):

HOLD = maintain the current power level

ACTION = change power (up or down) to a new level specified by the operator

OFF = no computer control (both software and the stepping motors were disabled).

Thus, the computer control was designed to be very much like the automatic pilot found on commercial airplanes. The computer could not do a startup (takeoff) or a shutdown (landing), but it could fly straight and level, and it could ascend or descend to a new level. The benefit to the production reactors was that every control action was made in such a way as to equalize the power distribution within the reactor core since the data to do this was at hand. This ensured the most productivity (within the hydraulic limits) and the most uniform product. Computer control had the additional benefit that it checked all of the requirements for safety circuits and hydraulic limits before it would raise power. Closed-loop computer control was used for about 90% of a reactor production cycle and proved to be a most diligent “operator”.

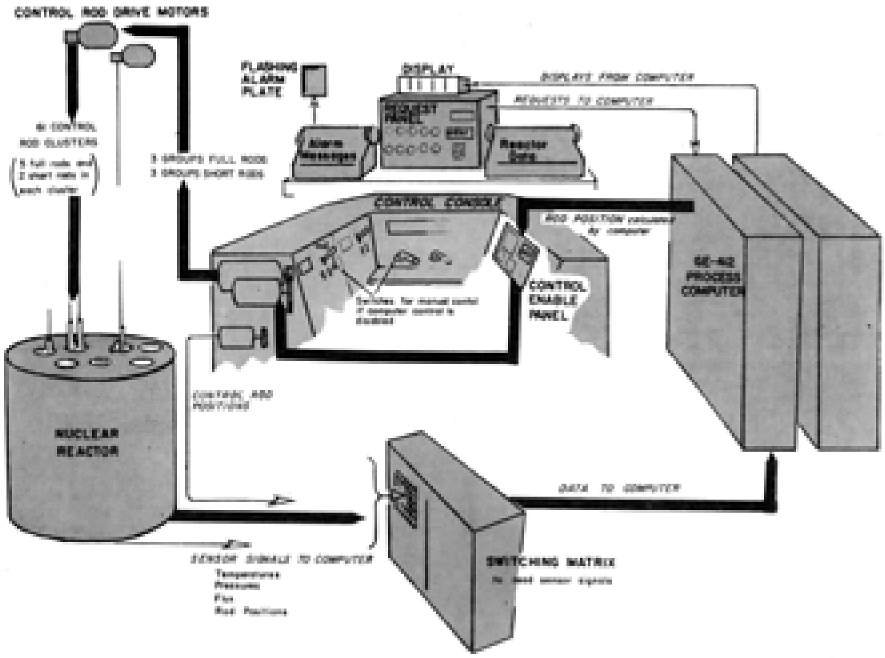


Figure 3. Automatic control of a nuclear reactor using a computer

1974—Dual Safety Computers

Ten years after the initial installation, on-line computers were much smaller and much cheaper. The installation at each SRP reactor was upgraded to a four-computer system. Two computers took over reactor control functions; these computers were essentially unchanged, but with one serving as a backup, yielding an availability of 99% of the time. The other pair was called safety computers and replaced the original mechanical safety circuits, which had hundreds of individual adjustments on one wall of the control room. Each computer monitored the flow signals from half of the 600 reactor assemblies and the temperatures signals from the other half of the assemblies. This arrangement provided either flow or temperature monitoring for the coolant to each reactor position, even if one computer was off-line. They were programmed by the Reactor Technology Group to be a safety circuit, capable of shutting down the reactor in one second if the safety limits on flow or temperature were exceeded.

1980—Automatic Backup for Fast Shutdowns

There had always been a concern that the safety rods of the fast shutdown system might fail to drop into the reactor for certain highly unlikely circumstances. For example, a severe earthquake might displace the reactor core or distort the safety rod guide tubes. A backup shutdown system was installed that could inject a liquid neutron absorber under high pressure. The system was manually activated by the control room operator. But by 1980, the safety computers had proven to be reliable enough to serve as a backup for operator action. The logic for the software was simple. The safety computers would monitor all circuits that could initiate a fast shutdown. If a fast shutdown was called for, and if reactor power didn't decrease by at least half in a few seconds, the safety computers would fire explosive valves to inject the backup system liquid into the reactor core. The plumbing for this system was made redundant, to ensure full effectiveness even if only one safety computer was operating.

1982—Automatic Diagnosis of Multiple Alarms

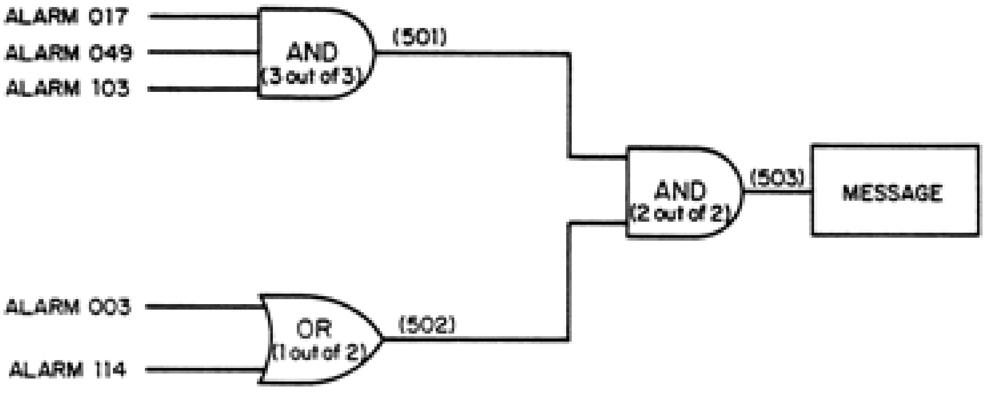
If the previous application was a simple extension of existing technology, the diagnosis of the plant alarms definitely was not. Our review of the reactor accident at Three Mile Island (TMI) led to some new research, which in turn led to a system to diagnose multiple alarms. It was called the DMA system and was installed at all SRP reactors.

The main finding of the TMI accident study was that the plant operators got so many alarms that they were overwhelmed mentally. They got over 100 alarms in the first 5 minutes of the accident. Some indicated minor problems, some simply reported a change of state, and some were very important. By the time the important ones came on, they were buried in the confusion.

SRP also had procedures to deal with individual alarms and combinations, but not 100 alarms in 5 minutes, which we concluded was also possible at our reactors. We decided to take the diagnostic steps in all those procedures and put them into "fault trees"—the same type of fault trees that General Motors was starting to use in their automotive shop manuals. This was a one-year task. Then we worked out a new way to store these fault trees in a computer in "tabular form" (see Figure 4).

This was the breakthrough needed, for now we could use ONE computer program to do all of the fault trees as each new alarm signal came in. The computer could easily keep up, and it could determine which fault tree had gone the farthest and identify the source of the trouble (see Figure 5). This was important stuff. The online computers would automatically diagnose what was wrong and tell the operators in plain English.

The concept of having a computer do a diagnosis based on symptoms and stored knowledge was known as an expert system. Many research groups were working on expert systems, but ours was the first that converted the logic trees to tabular form. This simplified programming and gave a simple way (using checksums) to validate any changes to the stored knowledge. After all, if you were going to let a computer diagnose something, you wanted to be sure it was right.



Decision	Number of Inputs	Number Required	Input Identification	Message
501	3	3	017, 049, 103	NO
502	2	1	003, 114	NO
503	2	2	501, 502	YES

Figure 4. Sample alarm logic tree and decision table

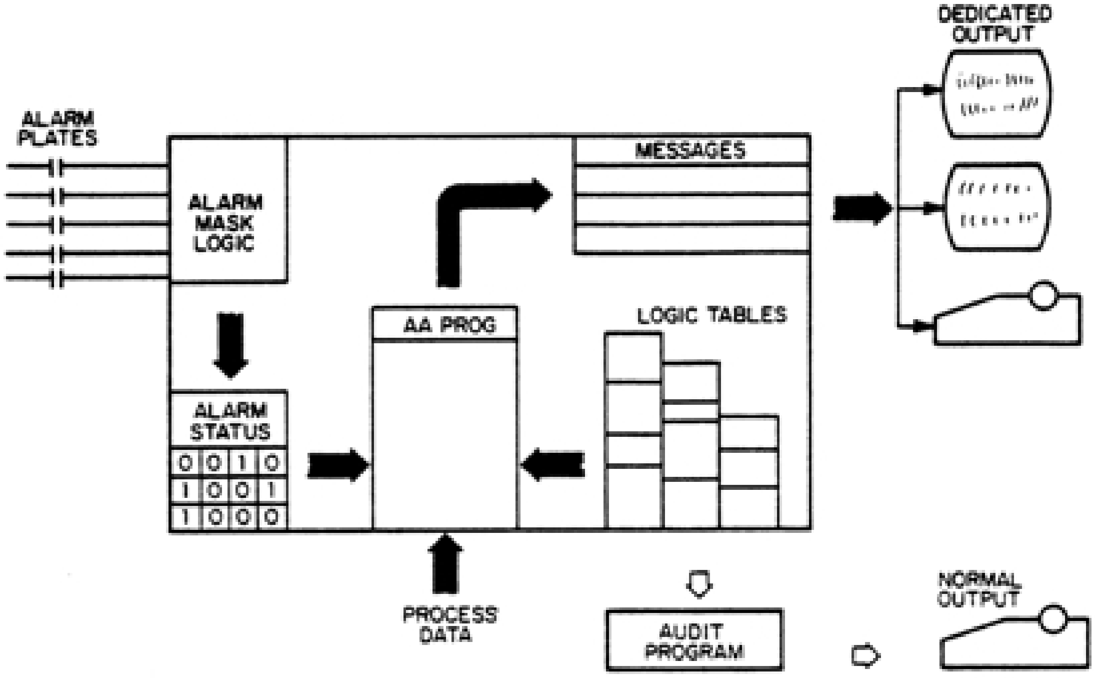


Figure 5. Data flow for alarm analysis

Biography

Kris L. Gimmy is a chemical engineer from Washington University in St. Louis, Missouri. During a 33-year career with Du Pont, he worked in all three divisions associated with nuclear reactors at SRS. He worked as a shift supervisor in Reactor Operation, as technical support in Reactor Technology, and in reactor safety research at Savannah River Laboratory. He also served as a consultant to the U.S. Nuclear Regulatory Commission.

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Reactor Safety Management Systems for the Savannah River Reactors

Ben C. Rusche

Abstract

Reactor safety management systems for the Savannah River heavy-water-moderated production reactors were established to ensure that public and employee health and safety were protected while meeting very demanding production objectives. Operational approaches and philosophies to achieve both safety and production objectives were developed by Du Pont based on experience at the Hanford Works and earlier experience in hazardous chemical and explosive manufacturing operations elsewhere in Du Pont Company. These systems were formally approved by the Atomic Energy Commission (AEC) as meeting the stated objectives and serving the national interest. This paper describes the evolution and function of these processes and practices and briefly chronicles the excellent performance at the Savannah River Plant (SRP).

Reactor Safety Management Systems

Origin

In the 1940s, after Enrico Fermi and his colleagues had achieved the first controlled nuclear chain reaction at the University of Chicago's Stagg Field, the promise for producing enormous amounts of energy and converting plentiful U-238 to Pu-239 was confirmed. Concurrently, the chemistry of uranium and the transuranic elements (e.g., neptunium, plutonium) were investigated by Glenn Seaborg (and others in later years) and his colleagues at the University of California. With understanding the chemical properties of these exotic elements, it became evident that the path to substantial quantities of fissionable material for power production or weapons was more practical and efficient by converting U-238 to Pu-239 in a reactor using the naturally occurring mix of uranium isotopes followed by chemical separation. The earlier processes of separating U-235 from natural uranium electro-magnetically in Calutrons (located at Oak Ridge) and later by the gaseous diffusion process (also located at Oak Ridge) continued to be used in the transition using natural uranium. Realizing the preferred course to plutonium production was through converting uranium in a nuclear reactor led to the request that the Du Pont Company undertake the task to design, engineer, and operate a major manufacturing complex at Hanford, Washington, to produce plutonium. To this task Du Pont assigned many of its best and most highly regarded engineers, physicists, chemists, and technical staff.

Those activities were important precedents to developing and implementing the system that became the pre-cursor for safety management systems for the Savannah River heavy-water-moderated reactors, which began operation in 1953. These reactors became a major source of U.S. plutonium production and the leading source of tritium for the free world. In later years, the U.K. and the Soviet Union developed substantial capabilities as well. First the Hanford and later the Savannah River reactors were designed and built by Du Pont with input and participation of some of America's most outstanding scientists and engineers.

Management Systems

Reactor safety management systems at SRP were prescribed by nuclear safety control procedures. These documents were authorized and approved by the senior management of the Atomic Energy Division (AED) of the Du Pont Company, and constituted the exercise of Du Pont's commitment to protect public health and safety while maximizing production rates and product quality. The activity was carried out by two divisions of the AED, the Manufacturing Division and the Technical Division. Upon completion of construction by the Construction Division, the Manufacturing Division operated and maintained the facilities. The Technical Division ensured that the best available technical designs were developed and tested, and that parameters for managing and controlling the operations were consistent with the procedures.

The material requirements of the AEC changed as weapons technology and national security needs evolved. The designs of the individual reactor charges were virtually tailor-made or adapted to meet specific product requirements even though the physical features of the SRP reactor systems changed only occasionally over the years.

It is the purpose of this paper to briefly describe the reactor safety management systems that allowed the AEC's objectives to be carried out safely and efficiently.

Organizational Relationships and Detailed Descriptions

The Atomic Energy Commission (AEC), established in the 1940s as a successor to the Manhattan Project, carried out its functions through several divisions. Of particular relevance is the Production Division responsible for manufacturing nuclear weapons materials. The AEC also established an Advisory Committee on Reactor Safeguards (ACRS) to provide independent review and advice on the adequacy of AEC reactors and their operating contractors (e.g., Du Pont at Savannah River) to protect the public and employees. In August 1964, the ACRS reviewed the SRP reactor operations and raised questions on terminology and on the safety bases for operations. In response to these questions, an active dialog followed for several months.

Reactor Safety Management Systems Principles and Terminology

The following material, largely extracted and condensed from a letter from J. W. Croach of Du Pont to R. C. Blair of AEC, Savannah River, dated September 24, 1965, presents the features of the Du Pont reactor safety management systems (Croach 1965).

Introduction

We believe it is important for the ACRS and for all personnel who have an interest in reactor safety at SRP to understand our principles of management controls and to appreciate the significance of the terms we use. It is especially important for the members of the ACRS because approval for new operating modes at Savannah River is sought on the basis that we will establish limits of operation in accordance with our standard practice; approval is not sought for specific power levels or other numerical parameters of operation.

Savannah River reactors are operated under a system of management controls that are designed, above all, to ensure safety, but also to permit the achievement of high performance levels. We believe reactor safety is best ensured by the multiple defenses of a sound process, reliable facilities, and responsible operation by qualified personnel. Perfection cannot be attained in any one of these; we believe the risk of a serious accident is minimized by incorporating multiple, independent protective features in the process, in the equipment and instrumentation, and in the management of operation. Most of the system of "defense in depth" is beyond the scope of this discussion (but underlies the entire philosophy). The basic features of the management controls that govern safe operation are discussed briefly in the following section.

Objective

It is our objective to operate the reactors under conditions where: (1) the limiting hazards of operation have been identified and evaluated, (2) regions of operation with acceptable risks have been established and duly authorized, and (3) methods of operation have been agreed upon and approved in advance of operation.

Principles

There are no generally accepted methods for quantitatively weighing and specifying risks. In general, a "risk" combines the concepts of potential damage to the reactor or its components and the likelihood of such damage. Associated with damage is the risk of releasing radioactivity that could be hazardous to the public. What constitutes an acceptable risk depends on technical analysis, management experience, and judgment. It is recognized that zero risk is a desirable limiting state but can only be achieved in practice by not operating the reactors.

For a given operating mode, the condition of the reactor at any time is described by values of measured or calculated variables that characterize the performance of fuel assemblies and of the entire reactor. These variables include such quantities as temperatures, coolant flows, heat fluxes, radiation fields, and thermal and mechanical stresses on the reactor structure. Prior analysis of operating characteristics and experimental data establish the values of critical operating variables at which actual damage or other undesirable consequences would occur in the reactor. Safe operation demands that these critical variables be rigorously controlled. There are usually several potentially limiting conditions that must be guarded against in operation, and any one of these might limit operation at a given time. For instance, depending upon the cooling water temperature and the radial flux distribution, a particular fuel loading might be limited by one or the other of the following: (a) boiling instability in some subchannels of the coolant passages in fuel assemblies, (b) film boiling burnout on the most vulnerable surfaces of the fuel, or (c) boiling the moderator outside the fuel assemblies.

For a particular critical variable, the principles of safe control employed at SRP involve the following:

1. Analysis to determine what value of the variable will yield actual damage—or what range of values has a high probability of damage. (Real Limit)

2. Agreement upon the value at which the probability of damage or harmful consequence is acceptably low. (Technical Standard Limit)

3. Designation of a safety margin to be maintained between the Technical Standard Limit and the range authorized for normal operation. The margin is selected to provide an acceptably low risk that equipment failure, operating error, or process fluctuations will result in damage. (Minimum Margin; Operating Limit)

4. Methods of operating the reactor and of measuring or calculating the critical variables are agreed to in writing in advance of operation. (Standard Operating Procedures)

5. Operation is continually surveyed and audited to ensure that the operation is in accordance with the intended control methods and that risks associated with the particular critical variable do not exceed those anticipated when the methods were specified.

6. Control methods and values are modified to reflect pertinent operating experience, improved equipment and instrumentation, new technical data, or changes in operating modes.

Terminology

Important terms used in the system of management controls are discussed briefly.

Real Limit. This term is frequently used in the discussion of a potentially limiting phenomenon that is capable of causing damage to the reactor, such as melting fuel. When a critical variable that governs the phenomenon has a value at which actual damage is expected or is highly probable, the value may be referred to as a "real limit". Sometimes the term is used to designate a particular value of a variable where there is an abrupt transition in the nature of the associated phenomenon (such as the onset of boiling) and where large uncertainties enter into the attempt to extrapolate the subsequent course of events. In any case, the probability of associated damage is high.

Frequently, the "real limit" is more appropriately regarded as a band of unacceptably high risk of damage. The probability of damage approaches unity and the magnitude of possible damage increases as the value of the variable approaches one edge of the band, while at the other edge of the band the risk borders on the acceptable—and in fact coincides with the Technical Standard Limit which will be discussed next. It is apparent that risks—both from the point of view of consequences and probability—can rarely be assigned definite quantitative values and that the selection of a boundary between regions of acceptable and unacceptable risks must be made through analysis on the basis of judgment and experience.

Technical Standard Limit. This is a formally approved and authorized limit that is not to be exceeded. It states the value of a critical variable that separates safe operation from operation where undesirable consequences may occur. The limit is selected on the basis that, at this value and for less extreme values, the risk is acceptably low. The analysis on which the limit is based includes a conservative allowance for uncertainties in the calculations, the accuracy and applicability of the data, and, if significant, an allowance for the accuracy with which the critical variable can be measured (or calculated from measurements).

If a Technical Standard Limit is exceeded, the condition must be corrected immediately. A special investigation and the preparation of a report to management in Wilmington are required. The objective of our system of management and controls is to maintain operation within the limits set by Technical Standards.

Minimum Margin. When a Technical Standard defines a limit that is critical and potentially limits reactor power, it specifies a Minimum Margin. This is defined as the minimum separation between the Technical Standard Limit and the Operating Limit. The Operating Limit may provide for a greater margin than the Minimum Margin. The Minimum Margin is established on the basis of technical information and a conservative evaluation of the consequences of abnormal operation and/or credible accidents; the bases for selection are specified in the Technical Standard. The purpose of the Minimum Margin is to provide factors of safety that will maintain a low risk of damage if any of the abnormal operating conditions and/or credible accidents described by the Technical Standard occur.

Operating Limit. In general, the operating departments specify Operating Limits on the basis of process knowledge, operating experience, available control instrumentation, and expected modes of operation. The choice of an Operating Limit may take into account factors other than safety, such as economy and operating convenience. One important objective in the selection of an Operating Limit for a reactor variable that has safety implications is to provide an adequate margin so that process fluctuations have a vanishingly low probability of exceeding the Technical Standard Limit. When a Minimum Margin is specified by the Technical Standard, it may be judged to be an adequate safety factor, or an additional margin may be specified. The Operating Limit indicates the highest level of authorized operation.

Standard Operating Procedure. The Standard Operating Procedures are the embodiment of the principle that operation of the reactors is to be carried out by methods that have been agreed to and approved in advance. These procedures specify in detail how the reactors are to be operated, what data shall be recorded, and what action must be taken to cope with unusual or emergency conditions. The Operating Procedures contain detailed limits and rules designed to keep the critical variables within the limits and intent of the Technical Standards and Operating Limits.

Technical Specifications. The Technical Specifications represent the instrument of administrative control of reactor operation by the AEC and, for SRP, are administered by the Savannah River Operations Office (SROO). Conformance with Technical Specifications is achieved by the requirement that Technical Standards must be equally restrictive or more restrictive than corresponding Technical Specifications. Violation of a Technical Specification requires a special investigation and the preparation of a report to management in Wilmington and a report to SROO.

For those who might desire more detail, a later version of Nuclear Safety and Control Procedures (1976) may be found in the last reference. Also, see Millison (1991), which contains a compilation of precedents to the final version of Technical Specifications utilized by Westinghouse Savannah River Company (WSRC). It is apparent that the level of detail increased and the scope broadened somewhat. Even so, the concepts and approaches for assuring the safety of the SRP reactors remained consistent with the earlier version.

Conclusion

The exceptional combination of conceptual approach in Reactor Safety Management Systems, physical design of the reactor systems, operational procedures, safety equipment, prior and concurrent technical design input, and large-scale verification (i.e., an extensive quality control and assurance activity) led to achievement of the AEC's and Du Pont's safety goals while increasing production rates (i.e., thermal power) by a factor of more than four over the operating life of the facility. For all operations at SRP through September 1998, the two highest hypothetical annual effective radiation doses to the maximally exposed individual in the public because of atmospheric releases of radionuclides from SRS were 11 mrem in 1955 and 14 mrem in 1956. All other annual radiation doses through 1998 were below 10 mrem. The current DOE (1990) and EPA (1989) annual limit is 10 mrem. This limit did not exist in 1955-56 (Carlton 1988).

For releases from SRP to drinking water sources for the entire operational period, the annual public exposure value has not exceeded 1 mrem—the maximum value was 0.8 mrem at Port Wentworth (Savannah, Georgia) in 1963. The national standard is 4 mrem adopted by DOE in 1990 and EPA in 1977. Thus, both the safety and production objectives were met (Carlton 1988).

The efficacy of the Safety Management Systems to commercial power reactor processes was recognized in the mid 1960s when a task force of experienced people from the AEC complex were formed to recommend regulatory and control processes for commercial power reactors. Marvin Mann of the AEC, a former Du Pont Savannah River Plant technical manager, formed the group which included A. A. Johnson of Du Pont, SRP; Herb Kouts of BNL and the ACRS; Joe DiNunno, AEC Division of Licensing; and a representative from Los Alamos National Laboratory. (The author served as staff to A. A. Johnson.) Out of this effort emerged the prototypical technical specification system as the licensing basis for acceptably safe operation of commercial reactors. As a final footnote, the author was appointed the first Director of Nuclear Reactor Regulation in 1975 when the Nuclear Regulatory Commission was established by Congress. The experience and relevance of the Du Pont commitment to reactor safety and the writer's personal participation in that evolution under the direction of A. A. Johnson were certainly factors in that selection.

On this note the story is closed, and the operation of reactors at Savannah River became history when the last remaining operating reactors were shut down in 1988.

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Biography

Ben Rusche joined the Du Pont Company at Savannah River Plant in 1953 with a BS in Physics from Tennessee Polytechnic University with later study in Nuclear Engineering at Georgia Institute of Technology and Stanford University. After 20 years at SRP, the Nuclear Regulatory Commission selected him as the first Director of Nuclear Reactor Regulation in 1975. He returned to Du Pont two years later as Corporate Director of Health and Safety. In 1980, he went to Washington as Special Assistant for Policy and Programs to Energy Secretary Dr. James B. Edwards. In 1984, he was appointed Director of the Office of Civilian Radioactive Waste Management (Assistant Secretary) for the Department of Energy in President Reagan's presidency. In 1988, he became Senior Vice-President in the Law Engineering Company in Atlanta. Since retirement in 1996, he consults in energy and environmental matters through his own company, Management and Technical Resources, Inc., located in Columbia, South Carolina.

Experimental Thermal-Fluids Program In Support of Reactor Operations

David Muhlbaier, Sam Mirshak,
Vascoe Whatley, and Elwyn Wingo

Abstract

Production at the Savannah River Site (SRS) was authorized in 1950 to produce nuclear materials for weapons. The Cold War was in full swing, and the Soviets had developed thermo-nuclear weapons. Our goal was to safely produce as much material as possible as quickly as possible. The mission was clear.

A key link in that long chain of events required to maximize production was to increase reactor power. To achieve this, a good understanding of the controlling phenomena was needed as well as a strong technical basis. The experience at Hanford provided some of the information needed to design the reactors. But these reactors were totally new and large machines that needed on-going technical support. Accordingly, a research effort was started (initially at Columbia University before any facilities were available at SRS). The research and experimentation were soon moved to SRS as the plant was constructed. The heat transfer and fluid mechanics work was assigned to "Pile Engineering" in the Savannah River Laboratory. ("Pile" was the original name for "reactor". For many a young engineer on his first assignment, the name conjured all kinds of meanings.) The research effort, along with hardware changes in the reactors, allowed the power to be increased by more than a factor of six. This document describes some of the key technical advances stemming from the experimental effort along with some anecdotal accounts by some of the people involved.

First Experiments

The first experimental facility at SRS was CMX (Corrosion Mockup eXperimental). It was built on the banks of the Savannah River to determine what water treatment was needed to prevent fouling the reactor heat exchanger tubes. The reactors were cooled with heavy water, which dumped its heat to the river water through large heat exchangers. Frequently, heat exchanger tubes will foul when cooled by river water. The reactors were initially designed with large water treatment plants to thoroughly treat all the river water used for cooling (approximately 90,000 gpm per reactor). This was a large and expensive treatment process, and it was desired to know the minimum treatment required to reduce cost. Accordingly, heat exchanger fouling experiments were initiated at CMX. Within a few months it was determined that no treatment was necessary. The abrasive characteristics of the river water scoured the tube surface and prevented significant fouling. Any fouling that did occur could be cleaned with oxalic acid. As a direct result to the CMX tests, one of the largest cost savings ever (for the era) was achieved.

A key element in achieving high reactor power (production) stemmed from the reactor design, which provided much of the instrumentation and to monitor the performance of every reactor fuel and target assembly. The design of the initial fuel assembly consisted of four slug columns in ribbed channels in one housing unit called a "quatrefoil" (Mark I assembly). The assemblies were installed in the reactor and rested, covering a monitor pin in the bottom of the reactor. The monitor pin contained four pressure taps (pressure averaged) to indicate assembly flow and four thermocouples to measure the effluent temperature from each slug column. When R Reactor was started (the first), it was found that the flow and temperature monitoring was erratic. Methods were proposed to improve the monitoring, but they had to be tested before making any changes to reactor hardware. Test results did show that hardware changes could correct the problem. Subsequently, all new reactor assembly designs (and there were many) were tested to ensure accurate monitoring of each reactor assembly. Accurate flow and temperature measurements were critical, especially as the power went up, because the signals were monitored continually to maintain safety margins and maximize production.

Vibration Wear Problems

Cladding failure of the slugs was an early reactor problem. Because of clearances between the channel ribs and the slugs, which were required to load and unload the slugs in the channels, slugs vibrated against the ribs. If the vibration were severe enough, the slug cladding could be penetrated. The hot uranium and hot coolant could react to produce uranium oxide. Two immediate problems were associated with cladding failure. First, the slug would swell because of the increased volume of oxide, such that channel flow would decrease and possibly overheat other slugs in the channel. Second, fission products would be released into the bulk reactor coolant/moderator, causing excessive radiation in external piping and penetrating the first barrier to the environment.

The vibration problems were reduced, but never eliminated, by reducing clearances between ribs and slugs and by reducing inlet flow velocities at the tops of the slug columns. The CMX work consisted of long-term flow testing to characterize any wear and the effect of changes in assembly design.

Moderator Circulation Problems

Reactor instrumentation indicated that “hot spots” or areas of poor circulation occurred in the bulk moderator inside the reactor tank. A full-size mockup of one-sixth of a reactor tank, called a crossflow tank, was constructed to study the circulation problems. One wall of the crossflow tank was constructed to allow visual observation inside the tank. Dye could be injected into the flowing water at the point of interest, and the flow path of the dye could be observed and its velocity determined by time of transport.

A large area of poor circulation was found in the tests. A special assembly called a jet-tube sparger (sparjet) was designed and installed in the crossflow tank to improve circulation in this area. The “jet-tube” portion of the assembly consisted of coolant flowing downward as in a fuel assembly but discharged upward through nozzles at various elevations along the length of the assembly. The upward, high-velocity flow promoted circulation in the “dead” spot. Six of the assemblies were placed in each reactor in the symmetric location determined by the single assembly in the crossflow tank. The sparger portion of the assembly was also used to provide the means to inject nuclear poison into the moderator for rapid reactor shutdown in the event of failure of the normal and emergency shutdown systems.

Increased Heat Transfer Surface Area

Reactor power (production) was severely limited by the then allowable maximum surface temperature and surface area available for heat transfer within the fuel elements. Programs were implemented to increase each and provide a sound technical basis. One of the first steps was to make the slugs hollow instead of solid. This step doubled the number of coolant flow channels. Previously the flow in each channel had been calculated. But with the increased complexity and higher assembly power, it was necessary to measure the flow distribution. This would allow reduction in the uncertainty allowances used to calculate the power limits and to establish a better technical basis.

Methods were devised to install pressure taps near the top and bottom of each coolant channel. By restricting the flow to one channel at a time, the flow and differential pressure relationship could be established. Then when the full assembly was operated, the flow split among the channels was determined. This information was needed input for the power limits calculation.

Failure is a natural part of success. In another attempt to increase the surface area of fuel elements, a bundle of five thin flat plates with ribs was designed and built. Hydraulic tests at CMX were required before the element could go into the reactor. While attempting to load the plates into a special housing tube, the plates stuck and would not go in or out of the housing. The design was subsequently abandoned; it was commented that we were trying to put a square peg in a round hole.

The next attempt at fuel elements with extended surface areas was the concentric tubular assembly (nested tubes). This design was the result of a newly developed co-extrusion process. The design was so superior that it was used from then on. The technology for measuring the flow distribution and temperature monitoring was already developed, so the transition to the new design was comparatively easy. The rest is history.

Two-Phase Flow

Early on, management wanted to know about the conditions necessary for the onset of flow instability. (Flow instability is an excursive process whereby excessive power of an assembly causes two-phase flow to develop and the flow rate to drop quickly to near zero. Once the flow decreases, overheating and melting can occur if the power is not immediately reduced.) That initial investigation was the start of an enlightening experience in two-phase flow of steam-water mixtures for many engineers. Some quick and dirty experiments were devised. A 14-foot-long tube was heated by two Lincoln welding generators hooked in parallel and connected for direct resistance heating. Flow through the tube mocked up the flow in one channel of the fuel assembly. CMX was chosen as the test site.

The tube was brought up to power, and then the flow was gradually throttled until the outlet temperature of the coolant reached the boiling point. The flow then decreased catastrophically as the pressure drop for two-phase flow was significantly greater than for single-phase flow. We had achieved our first flow instability. Power was maintained on the tube, and it became glowing red hot and warped and wiggled like a snake. If that tube had been a fuel assembly, it would have melted and released fission products to the moderator coolant and possibly to the environment. The major concern was reactor safety and melting of the fuel. Such tests were used to help determine reactor power limits.

Heat Transfer Laboratory

The nameplate rating of the reactors was 378 MW. This was based on a Mark I fuel assembly with a maximum surface temperature of the aluminum cladding of 80^{0}C. We carefully calculated power limits based on not exceeding an 80^{0}C surface temperature, including hot spots on the surface of the aluminum cladding and a maximum central metal temperature of the uranium of less than 600^{0}C. These limits were based on concerns about aluminum corrosion and uranium swelling from phase changes.

New corrosion studies showed that in the high-purity coolant, the temperature was very conservative. We set out to find out what the limit on the heat removal capability of the fuel was, since this was directly proportional to producing nuclear materials. A Heat Transfer Laboratory was built in Building 773-A to study assembly cooling phenomena and power limits. We determined that surface temperatures of the aluminum could be allowed to be consistent with nucleate boiling. However, we were defining the maximum heat fluxes from the assemblies without getting film boiling. Film boiling was referred to as the hot stove effect. If film boiling occurred, the surface would be blanketed with steam essentially resulting in adiabatic heating of the fuel and subsequent fuel melting. Several rigs were built to determine the maximum heat flux without incurring film boiling as a function of pressure, subcooling, and coolant velocity. Film boiling would essentially lead to melting the fuel surface and burnout. A burnout safety factor was born and referred to as the Burnout Safety Factor (BOSF), the ratio of burnout heat flux to operating heat flux. The data were correlated in an equation using the power of a slide rule, now practically extinct.

In one experiment while trying to get to the burnout heat flux, we kept reducing the subcooling and unknowingly got two-phase flow in the downstream piping. The glass tube that formed the annulus around the heated tube, which was glowing red-hot, ruptured, and, although it was encased in a “Plexiglass” housing, allowed steam to escape. We were almost trampled by one of our larger managers who was escaping from the “steam explosion”. At the next management meeting, the Laboratory Director pointed out with some indignation that unusual occurrences sometimes happen in the presence of the foreman.

We developed a correlation for the effect of spacer ribs to be used in conjunction with our burnout correlation. In an attempt to improve the precision of the heat transfer correlation, subsequent researchers developed a modified correlation. Unfortunately, this correlation was used with the earlier correlation on the effect of ribs on the burnout heat flux, which eliminated conservatism in the analyses. In some subsequent very-high-flux charges, some slight melting occurred along some ribs. Fortunately, the fuel exposure was very low, and no significant radioactivity was released.

The heat transfer experiments had established higher operating limits for the fuel. Engineering developments and design had significantly increased the assembly surface area. Together they provided higher assembly power by more than a factor of six. Reactor modifications were then able to take advantage of the higher power limit.

Other activities as an extension of the Heat Transfer Laboratory were related to PAR pond studies. A small lake behind C Reactor was built to determine the optimum way to introduce hot water into PAR Pond, which was then in the planning stage. Management wanted to know the effect of the depth of draw-off on the outlet temperature on an urgent basis as design of the lake was proceeding. The engineer and technician went to Central Shops salvage yard and found some galvanized ventilation pipe and two swivel joints and headed out to the pond. They took off most of their clothes, went into the pond, installed a variable draw-off and were collecting data the next day. A diesel generator and pump were operated to support the experiment. They requested equipment operators to watch it that night. The next day the operators said alligators were crawling out of the lake and over the dam. It was jointly decided that the equipment didn’t need surveillance at night.

Airborne Activity Confinement System (AACS)

When the reactors were designed and built, they contained a once-through ventilation system with essentially no filtration of the exhaust air. This meant that in the event of a significant reactor accident with fuel melting, large amounts of radiation could escape the building and contaminate the surrounding area. Calculations showed that such a postulated accident could also release large quantities of steam. A filtration system was wanted for the reactor ventilation system to significantly reduce radioactive releases in the event of a reactor accident. Tests of standard filtration systems showed that particulate filters could in fact remove more than 99.9% of all contaminants except iodine, tritium, and noble gases.

However, they could not survive for long in the presence of wet steam. Further testing established that a demister immediately upstream of the particulate filter would knock out enough water particles to prevent failure of the particulate filters. A large part of the required filtration system was thus defined.

No industrial process was available at the time to capture tritium or noble gases. However, it was known that iodine could be captured on activated carbon. Testing commercially available carbon beds showed that none of them could meet the requirement for 99.9% removal efficiency. Testing was undertaken to determine the cause of the low efficiency and to correct the deficiency. It was determined that a one-inch-thick bed of coconut-shell-activated carbon could meet the requirement. It was also determined that the iodine would pass along the interface between the carbon and the metal container wall with somewhat less adsorption efficiency. Redesign of the container frame to include baffles at the interface solved the problem. We then had a workable design for the reactor exhaust gas confinement system (AACS). The system was then designed and installed in all reactors.

But just because the filters were designed to remove more that 99.9% of the particulate and iodine, that doesn't mean that errors in the installation process couldn't defeat the objective. In-place testing was needed to verify the system performance. A standard test called a DOP test was available for particulate filters, and this was adapted for use in the exhaust gas confinement system.

A program was initiated to develop an applicable in-place test for the carbon beds. A substitute tracer material was needed because using iodine as a tracer would deplete the capacity of the carbon for the iodine and defeat the purpose of the test. A tracer material was needed that would adsorb, then desorb from the carbon but pause long enough to measure any leakage flow. Chlorinated hydrocarbons were proposed because they were expected to meet the requirements. Testing was initiated at CMX to verify the strategy and hypothesis. Several chlorinated hydrocarbons would meet the requirements, and the best was selected and demonstrated in a reactor. A 0.05% leak path was installed in one of the reactor filter compartments, and the test was used to determine if the known leak could be detected. The results clearly showed the leak path and the ability to detect leaks of less than 0.01%. We then had an exhaust gas confinement system that could meet requirements, and two in-place tests that would verify the fact.

The activated carbon bed testing method was totally new and was found applicable to the nuclear industry. The test was standardized for use in nuclear power plants and is still in use today.

Flow Oscillations

Very-high-neutron-flux charges were designed for the reactors in the latter part of the 1960s. This required fewer assemblies with much higher flow (>1000 gpm). The fewer assemblies were clustered near the center of the reactor and resulted in much higher velocities of coolant through the plenum. When the charge was operated, there were dramatic changes with time in the assembly flow rates. The flow varied up and down (oscillated) for no apparent reason and with a variable period. This required de-rating the design reactor power, and the reasons for the variation were investigated.

A full-scale mockup of the reactor plenum was built at CMX to investigate the cause of the flow oscillations. Cooling water was admitted to the assemblies through slots in the plenum sleeve. It was determined that the plenum was behaving like a large fluidic amplifier. That is, a small perturbation in one region of the plenum caused dramatic changes in fluid velocity in other parts of the plenum without changing the total reactor flow rate. The suspected cause of the small perturbation was vortex shedding, but this was never confirmed.

The assembly flow was sensitive to plenum local velocity because the inlet slots were located in the narrowest passage between plenum tube positions and, of course, this was the region of highest velocity. Hence, an increase in velocity perpendicular to an inlet slot caused reduced flow to the assembly. In fact, study of the data from previous operations revealed the same phenomenon had existed, only on a much smaller scale.

To reduce the effect of flow oscillations, several new potential designs for the inlet to the universal sleeve housing (USH) tube were tested. The USH passed through the plenum tube and held the fuel and target assemblies in the reactor. The old USH design contained slots that aligned with plenum sleeve slots. As a result of the testing, a new USH was designed that used 270 one-fourth-inch-diameter holes spaced uniformly around the sleeve. This had the effect of damping the variations in assembly flow with changes in fluid velocity in the plenum. That design was used permanently thereafter.

Starved Pump Test

The postulated loss of coolant accident (LOCA) was studied in the late 1960s. The LOCA is a very low probably accident characterized by an instantaneous pipe break with unimpeded discharge from both pipe ends. This accident would initiate emergency cooling (ECS) water injection into the reactor at considerably reduced flow to each assembly. However, the reactor would be shut down by then, so that only decay heat would need be removed.

Analysis showed that higher reactor powers could be achieved if credit could be taken for pump re-circulation. That is, given a LOCA and an empty reactor tank, the ECS may not be the only source of water. It was estimated that the pumps would return must of the ECS water to the plenum if they would in fact operate with what was termed a starved suction (an air-water mixture flowing into the pumps from the reactor). Preliminary tests were performed at CMX to characterize pump behavior under such conditions. Then, a reactor test was conducted in P Reactor to verify the expected performance. A great deal of instrumentation was installed to measure the system flows, pressures, and vibration at critical locations.

The primary test was started with five pumps operating at full power and one pump shut down to represent the pumping system with the broken line. The moderator level in the reactor tank was gradually lowered until the pumps began to aspirate air at which time the pumping systems began to make some noise. Typically, you could hear a quiet hum in the control room when the reactors were operating. As the level was lowered further, more and more air was aspirated into the pumps, and the noise became louder and louder. As the level reached the point of maximum air entrainment (just before loss of suction), it sounded like large rocks were being pumped in the system. The experienced operators were walking around the control room as on egg shells. Very little was said. The piping system was vibrating beyond anything anyone had seen, but it did not reach what was previously determined as critical. Once the pump suction was lost, the extreme noise and vibration vanished. The pumps then just re-circulated the water that ran into them by gravity flow and operated quietly. That was the result we had expected and wanted. It proved we could take credit for pump re-circulation in the event of a LOCA.

The New Heat Transfer Laboratory

The early 1970s brought more attention to severe postulated accidents. A new Heat Transfer Laboratory was built, and one of the first phenomena to be studied in more detail was flow instability. The new Heat Transfer Laboratory had 3 MW of installed and rectified power that could be used for electrical resistance heating of mockup fuel tubes, far more than was previously available with welding generators. This meant that full-scale mockups of one or two channels of a fuel assembly could be achieved with prototypic heat flux.

One of the first accidents to be studied was the pump shaft break accident (very low probability) in which there would have been a sudden reduction in coolant flow, which could drive the assemblies into flow instability if they were operating at too high power. The onset of flow instability would mean that fuel melting would very quickly follow if the power were not reduced immediately. This is a very fast accident that could quickly get out of control so that the reactor charge was operated such that flow instability would not happen even if a pump shaft break did occur. Experiments were initiated to better determine the power levels at which flow instability would be initiated given the accident.

The first experiment to study flow instability used a single-heater tube with an inter housing rod of fiberglass to form a single heated annulus. Electrical power at 120 volts and up to 30,000 amps was passed through the outer stainless steel tube to create the heat. The first test did not use full power but was sufficient to generate steam. The people involved had lots of experience with experimentation but no previous experience with flow instability. Precautions were taken to protect personnel against the unexpected.

The test was started, and the assembly brought up to power. When steady-state test conditions were established, a quick-opening valve was activated that caused a sudden reduction in coolant flow. Flow instability was immediately triggered. The engineers understood the flow instability process, but had little appreciation for its speed or violence. After the fact, it was recognized that the flow instability created extremely high pressures in the bottom of the assembly, which provided very large lifting forces on the inter-housing rod. The result was the rod broke its restraints and came out of the test assembly like a rocket. It rose to the ceiling, hit a wide-flange I-beam, was deflected, and created just a small dent in the roof. No one was hurt because the personnel were all in a shielded control room. However, the incident brought a new respect for high-powered experiments and pointed out the need to calculate expected results and forces to predict the consequences of a test. It was something that was done for the reactors, but now must be done for the experiments. It was called a learning experience.

Before and After the Three Mile Island Accident

The accident at Three Mile Island (TMI) marked a turning point in much of the history of nuclear reactors. In the late 1970s at SRS and before TMI, there was a drift away from experimental programs (because of their expense) and toward computational solutions. Experiments and calculations have always gone hand in hand. One cannot flourish without the other. But in a mature industry, it was believed that the experimental basis had been adequately established, and calculations could handle most new situations.

TMI changed that thinking. As the analysis of TMI evolved, more attention was given to low-probability accidents. Experiments were started in the mid 1980s to study the Loss of Coolant Accident (LOCA). The initial LOCA experiments studied the heat-removal ability at various flow rates down one channel of typical dimensions. The flow splits among the various channels of an assembly had previously been measured along with a conservative estimate of ECS cooling water flow rates. When this information was put together in a mathematical model that accounted for heat splits, the results showed there would be some melting of reactor assemblies in the event of an ultimate LOCA. This of course was not acceptable, and the reactor power was reduced about 25%.

The National Academy of Science was asked to review the results, and they wanted a more conservative power level, so the reactor power level was again reduced to about 50% of initial power. Furthermore, at about the same time, there developed a procedural issue for the startup of P Reactor after an extended shutdown. These conditions, along with an abundant supply of weapons material and the end of the cold war in sight, resulted in the shutdown of all the reactors to further study low probability accidents.

Multiple committees were established to review the limits system, and critics came from everywhere. Reviewers experienced with power reactors maintained that critical heat flux (film boiling and burnout on surfaces while maintaining flow) would be our limitation at high power and not excursive flow instability as our experience indicated. Hence, major programs were established to study film boiling and flow instability as well as ECS cooling conditions. The experimental programs were far more than could be accomplished in the Heat Transfer Laboratory, so contracts were let to B&W (Alliance, Ohio), Creare (Lebanon, New Hampshire), and Columbia University (New York City) for additional experimental work. Combustion Engineering was also contracted to produce a prototypic assembly for testing in the Heat Transfer Laboratory.

After about six years, the end results of the experimental and computational programs were verification of the flow instability criteria and establishment of new, more-conservative limits for LOCA where ECS cooling would be required. In the mean time, however, the Cold War came to an end, and DOE decided to shut down the reactors permanently.

Thus, a long history of experimentation in support of reactor operations came to an abrupt end. The program had provided direct support to both reactor operations and the technical basis for the computational program and the limits system. In the scramble to find other work or be shut down too, Heat Transfer Laboratory personnel scoured the plantsite looking for a new role. The laboratory name was changed to Thermal-Fluids Laboratory (TFL) to provide a more descriptive name for potential customers. Both large and small jobs were found for the transition, and now the laboratory is once again thriving with a broad base of customers. It is a tribute to the long history of excellence in the experimental program at SRS.

The success of the experimental thermal-fluid program was a direct result of the many excellent people who supported it. Credit belongs not only to the engineers, but also to the technicians, secretaries, and manager as well as other support services. The authors also acknowledge assistance in the creation of this paper from George Richardson, Jim Smith, John Steimke, and Vince Walker.

Biographies

David Muhlbaier

David Muhlbaier has a BSME degree from Drexel University and arrived at SRS in 1961. He worked for many years at CMX and was responsible for various experiments, including the development of the carbon bed leak test. He then became involved in two-phase flow experiments after the new Heat Transfer Laboratory was built. In 1980, he was assigned to the L-Reactor restart project and subsequently to Reactor Technology. He concluded his career as manager of the new Heat Transfer Laboratory beginning shortly after the initial shutdown of the reactors and during the time of extensive internal and external experiments designed to enable reactor restart.

Sam Mirshak

Sam Mirshak has an MSChE degree from Northwestern University and came to work at the Savannah River Site in 1953. He held progressive positions in research and development and management leading to Works Technical General Superintendent and Production General Superintendent for SRS. He was appointed Director of Research of the Savannah River Laboratory. In 1984, he was assigned Associate Technical Director for the Atomic Energy Division in Wilmington, Delaware. From 1987 to his retirement from Du Pont in 1989, he also served as Director of Nuclear Safety and Quality. Subsequently, he was a consultant for the Department of Energy and their national laboratories. Mr. Mirshak was the principal contributor to the heat transfer correlation that allowed a large increase in reactor power with demonstrated safety margin.

Vascoe Whatley

Vascoe Whatley has a BSChE degree from Clemson University and came to work at SRS Operations in 1956 after several short stints in SRS Construction. He worked much of his career at CMX in the experimental arena. He was the principal contributor to the hydraulic studies of new or proposed fuel and target assemblies and to the long-term vibration and wear issues with production assemblies. His experience included in-reactor testing. He also worked in isotope applications and then returned to CMX and subsequently to the new Heat Transfer Laboratory.

Elwyn Wingo

Elwyn Wingo has an MSME degree from Georgia Tech. He came to work at SRS in 1955 and worked as a test engineer at CMX. He was responsible for much of the experimental work that lead to the solution for inadequate moderator circulation and assembly flow oscillations, as well as various other experiments. He also managed work in the computational effort, principally in the arena of heat transfer and fluid mechanics. Other assignments included Reactor Technology, the Heat Transfer Laboratory, Long Range Planning, and Probabilistic Safety Analysis.

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Reactor Program for Increased Production Capability

James M. Morrison

Abstract

The Du Pont Company undertook the mission to design, build, and operate the then Savannah River Plant in 1950. A conservative design basis of 378 megawatts (MW) was established for the production reactors. As quickly as the reactors were placed in operation, a strategy was implemented for increasing their output. Numerous upgrades were installed in the cooling systems from 1956 through 1964 to increase power levels and production output. More process heat exchangers, larger piping, increased pump impeller diameters, new pumps, and PAR Pond with its pumphouse were added to increase cooling capacities, and blanket gas pressure was increased to allow higher operating temperatures. During the same period, a series of increasingly advanced fuel and target assemblies was introduced to improve productivity and take advantage of the increased power capabilities of the hydraulic systems. The Mark I fuel assembly was replaced in turn by the Mark VII, VII-A, V-B, V-E, and V-R as the standard for plutonium production. For tritium producing charges, the Mark VIII assembly was in turn replaced by Mark VI, VI-J, and VI-B assemblies. All these system and fuel upgrades were in place by 1964, and in 1967 C Reactor achieved a peak power of 2915 MW, more than seven times the original design power level. The extent and pace of this program represents an outstanding achievement by the thousands of people involved.

Introduction

The mission to design, construct, and operate the Savannah River Site (SRS) was undertaken by the Du Pont Company in 1950 in response to a request from the U.S. Government (Bebbington 1990). The urgency of this mission, in the context of the times accompanying the Cold War, was conveyed to Du Pont by President Truman. Nevertheless, Du Pont accepted the responsibility reluctantly, in part because they had no experience beyond their previous role in building and operating the Hanford reactors that would be directly applicable to the new facilities. The range of potential reactor types was quickly narrowed to a heavy-water cooled-and-moderated reactor, employing a secondary light-water system to remove reactor heat. With no such large-scale facility in existence to provide guidance, a design emphasizing versatility and a conservative design basis for power of 378 megawatts (MW) were established, and five reactors—R, P, L, K, and C—were constructed and in operation by 1955.

The output of a production reactor is directly proportional to the product of power and operating time multiplied by the conversion ratio (i.e., grams product per megawatt-day). Provided that time taken up in planned and unplanned outages is kept acceptably low, annual production of desired isotopes (e.g., plutonium and tritium) is thus directly related to the heat output of the reactor. With defense demands for special nuclear materials increasing rapidly, a program to increase reactor power and production capability was implemented as quickly as the reactors began operating. This program included various measures to increase the heat removal capability of both the reactor primary and secondary cooling systems. Concurrently, a robust program was undertaken to develop advanced fuel and target assemblies to match the ever increasing power potential of the cooling systems. These programs overcame many significant challenges in successfully increasing reactor power to a peak of 2915 MW, realized in C Reactor in 1967. A summary of the technical improvements involved in this outstanding achievement is presented in this paper.

Factors Affecting Production Capability: "What to Improve?"

There are a number of physics and engineering criteria that governed the power and production capabilities of the SRS reactors. Fundamentally, these are of two types: (1) basic reactor core design, including size, inventory and types of fissile and fertile materials, geometry, amounts of heavy water (D_2O) moderator and coolant, quantities of structural materials, and amounts of absorber materials used to control the nuclear reaction; and (2), the ability of the primary and secondary cooling systems to remove the heat of reaction. These two general categories were strongly interrelated in determining the output of desired products.

The core physics of a fuel charge determined its reactivity and productivity (i.e., conversion ratio in terms of grams of product made per megawatt-day [MWD] exposure or per gram uranium-235 fissioned). Using heavy water as both moderator and primary coolant contributed to a high productivity of Pu-239, tritium, or other desired products as a result of its very low neutron absorption compared to graphite or light water. Metallic fuels and targets were employed to maximize material loadings. Their use was feasible because the reactors operated at relatively low pressures and temperatures. The design of fuel and target assemblies, specifically coolant flow area and heat transfer area, was also a major factor in determining the ability of the primary coolant to remove heat generated within the assemblies by fission or neutron absorption.

The formation of fission products, or new elements formed when an atom splits into two elements, reduces charge reactivity. They add materials that absorb neutrons that would otherwise be absorbed in creating desired products; and, therefore, they limited the number of operating or "full power" days that could be obtained from a reactor charge. Operating time was also affected by:

- Time spent in scheduled outages to charge and discharge fuel and target assemblies
- Performing maintenance and repairs, and conducting tests of safety related equipment
- Time lost due to unscheduled shutdowns (real or spurious emergency shutdowns caused by abnormal conditions)

The design of the reactor hydraulic systems directly affected the ability to remove and dissipate the reactor power (i.e., the heat produced in the reactor core). The primary heavy-water coolant, also called "process water", passed through the fuel and target assemblies to remove the heat of reaction, exiting to the reactor tank. It then flowed to the circulating pumps, where it was pumped through process heat exchangers to transfer the heat to the secondary light water coolant, then back to the reactor assemblies. The light water was supplied by pumping either from the Savannah River or PAR Pond to 25-million-gallon retention basins in each reactor area. From there it was pumped through headers to the process heat exchangers, then back to either the river or pond, gradually dissipating the reactor heat to the environment. The heat removal capability of these systems was determined by the flow rates of both process water and light-water coolants, heat exchanger surface area and heat transfer coefficient, system pressures, and temperatures.

Strategy for Increased Production: "What was the Plan?"

The program to increase the production capability of the SRS reactors addressed and included all of the above factors in a comprehensive and systematic way. With the first reactor criticality achieved on the last day of 1953, reactor construction and startups continued until C Reactor was completed in 1955. By the end of that year, utilizing the installed reactor hydraulic system and the first fuel for plutonium production (Mark I) peak reactor power had reached 877 MW, well above the 378 MW design level. However, it was apparent that any further meaningful increase in power and production would require significant improvements to the hydraulic system, as well as developing advanced fuel assemblies with greater heat transfer capabilities to take advantage of increased reactor hydraulic power limits.

The strategy that was adopted contained three essential elements for implementation:

1. Enhance the process water and light water heat removal system capabilities to permit increased reactor power.
2. Optimize fuel and target assembly designs to increase productivity and take advantage of the steadily increasing hydraulic power limits.
3. Utilize an orderly approach to power ascension, by designating one reactor to “pilot,” or increase power in step-wise fashion ahead of the others, to minimize cost and safety risks.

Increased Reactor Capabilities: “What was Accomplished?”

Numerous changes were made to the reactor hydraulic systems, beginning in 1956 and continuing until they were essentially completed in 1964. These were planned and designed largely by Du Pont’s Wilmington Process Section and the Reactor Technology Section at SRS, with the Du Pont Construction Division responsible for actual modifications. At the same time, because of the close interrelationships between reactor hydraulic and fuel assembly power capabilities, a program was undertaken by the Technical Division to design a series of advanced fuel and target assemblies to match the ever increasing limits evolving from the reactor hydraulic programs. The Technical Division efforts were conducted primarily by the Savannah River Laboratory (SRL, currently designated the SRTC), and comprised both physics and engineering design as well as experimental verification of performance. The more significant enhancements resulting from the combined efforts of these production improvement programs are outlined in the following paragraphs.

The first fuel for the reactors, designed for plutonium production, was the Mark I natural uranium slug clad with a thin layer of aluminum. The cylindrical slugs were about 1 inch in diameter and 8.4 inches long. The aluminum housings, called quatrefoils, were composed of four nominally 1.5-inch-diameter hollow tubes with internal spacing ribs, arranged in a square pattern. Twenty slugs were loaded in each of the four tubes in a quatrefoil, which occupied one reactor position.

The initial power ascension program began in 1954-55 before any equipment or fuel type changes were made. With P Reactor acting as the pilot, power levels were increased in increments of about 13% to determine actual fuel and hydraulic system limits. Temperatures at key points in the reactor system (fuel assembly effluent, fuel cladding, fuel central metal temperature, reactor tank outlet temperature, etc.) were calculated and/or monitored, and safe operating limits were set. With each increase in P-Reactor power, fuel performance, reactor stresses, and other conditions were carefully evaluated before the other reactors were permitted to increase power. Various methods were used to enhance uniformity of individual fuel assembly power operation (i.e., reduce maximum/average ratios) to maximize total reactor power for a given fuel operating limit. These included radial spiking with special fuel assemblies (Mark VIII) containing enriched uranium (5% U-235) in the outer region of the core to increase reactivity and improve radial neutron flux shape and using partial (less than full active length) control rods to improve axial neutron flux shape. During this period, it became evident that, due to the low thermal conductivity of uranium metal, the progressively higher powers and operating temperatures were causing swelling and breaching the aluminum cladding in some Mark I fuel slugs, causing fuel failures. Reactor powers reached a peak of 877 MW by the end of 1955.

Power ascension continued in 1956. In July, L Reactor was made the pilot because of moderator turbidity (aluminum corrosion products suspended in the moderator) in P Reactor, and power ascension continued at a reduced increment of 8%. P Reactor again assumed the pilot role when turbidity decreased through a program of improved moderator chemistry. Flow zoning of the process water through the reactor core, tailored to fuel assembly radial power distribution, was initiated to improve available coolant use. Production of Mark VII fuel for plutonium production began in mid 1956 to replace the Mark I fuel and eliminate the central metal temperature limitation. Mark VII slugs were slightly larger in diameter than Mark I but had a central hole to allow coolant to flow both outside and inside the slug column.

The original reactor design called for plutonium production in the natural uranium fuel elements and supplemental tritium production in lithium-aluminum control rods. In the mid 1950s, however, requirements for tritium increased substantially beyond the incidental production capabilities of the control rods. Accordingly, special reactor charges were designed with tritium as the major product. These charges produced tritium in both the fuel assemblies and the control rods. The quatrefoils were loaded with a 3:1 ratio of Mark VIII enriched-uranium fuel slugs and lithium-aluminum target slugs. The fuel and target slugs were “stripe loaded” in the quatrefoils (i.e., in barber pole fashion) progressing down and around the four columns of each assembly. These charges were effective tritium producers. But, with the fuel elements having only 75% of the Mark I heat transfer surface, they operated closer to heat flux limits at any given reactor power.

The first major hydraulic system changes also began in 1956. Six more heat exchangers were installed in R, P, L, and K Reactors, piped in series with the original 6 (C Reactor was originally equipped with 12 exchangers). River water flow was increased by installing larger impellers in the Building 190 light-water pumps used to move cooling water from the 25-million-gallon retention basin through the heat exchangers in each reactor area. The increased light-water flow and heat exchanger surface area were effective in reducing process water temperatures and allowing higher power operation. Power ascension resumed in late 1956 in 60 MW increments. The combined effect of all the changes that had been made was to double reactor power, which reached a peak of 1380 MW by the end of 1956.

The next major system upgrades were begun in December 1956 in C Reactor and were completed in all reactor areas by 1958. These upgrades included:

- Replacing the six Byron Jackson process water (PW) pumps with higher flow, lower head pumps manufactured by the Bingham Pump Co.
- Increasing the diameter of the PW piping and re-piping the PW heat exchangers (HXs) parallel, rather than series, to accommodate the higher flow
- Installing even larger diameter impellers in the Building 190 light-water pumps

The combined effects of these changes were to increase PW flow by 75% and cooling water (CW) flow by 70%, greatly increasing the power capabilities of the reactor hydraulic systems. More advanced fuel assemblies were needed to take advantage of the increased power potential. For plutonium production, the Mark VII-A design replaced the Mark VII beginning in June 1957. The Mark VII-A fuel was designed for use with the new Bingham pumps. It was similar to the Mark VII but somewhat larger in diameter and with a larger central hole. It was designed for use with the largest quatrefoil that could be inserted through the reactor stainless steel semi-permanent sleeves. For tritium production, Mark VI series fuel elements were designed by SRL to replace the Mark VIII assemblies, beginning in 1957. The first of this series, Mark VI, was an assembly of thin concentric tubes, one tube containing fuel (high enriched, 93% U-235, uranium-aluminum alloy) spaced between two aluminum housing tubes, and an internal slug column of target material (enriched lithium-aluminum alloy). The successful introduction of the Mark VI design was pivotal in developing a series of completely tubular, extended surface area designs for both fuel and target materials that were more efficiently matched to the higher Bingham pump flows and replaced the older quatrefoil assemblies.

The net effect of all these changes in both fuel designs and hydraulic systems was a significant increase in reactor power. Peak power increased from 1380 MW to 2250 MW by the end of 1957 and to 2350 MW in 1958, while average power level increased by 400 MW in 1958 compared to 1957. In 1959 additional CW capacity was added with the completion of PAR Pond. PAR (acronym for P and R) Pond is a 2600-acre lake created by damming Lower Three Runs Creek, constructing a pump house, diverting R- and P-Reactor effluent CW to the Pond rather than the river via canals, and using the Pond to cool the effluent CW from R and P Reactors and recycling it back through the 25-million-gallon retention basins and heat exchangers. In this way much of the river water formerly pumped to R Reactor and P Reactor, which were situated farthest from the river, could be diverted to L, K, and C Reactors. The net gain realized from PAR Pond was an increase of 850 MW in power (total for all five reactors). Mark VI-J fuel replaced the Mark VI design for tritium production beginning in 1959 to obtain more favorable physics characteristics. The Mark VI-J also had a single enriched uranium-aluminum fuel tube, but the central slug column was replaced by a thin, hollow lithium-aluminum target tube.

Additional changes were made to the river and Pond CW systems in 1960 to increase CW flows still further. These included:

- Increasing impeller diameters of the 20 river water and 7 PAR Pond pumps
- Adding three more PAR Pond pumps
- Adding one new double capacity pump to each of the two Building 190 headers supplying water from the retention basin to the heat exchangers in each reactor area
- Constructing a new effluent ditch from P Reactor to PAR Pond

The combined changes increased the nominal CW flow rate to each reactor from 150,000 to 175,000 gallons per minute (gpm). As a result of these improvements in 1960, C Reactor achieved a peak reactor power of 2575 MW early in 1961.

The last project to significantly upgrade the power rating of the reactor hydraulic system was carried out in 1962-63. This project increased the helium blanket gas pressure from slightly above atmospheric to 5 psig. This increase had the effect of increasing saturation temperatures and safety-related temperature limits throughout the system, such as fuel assembly effluent, pump cavitation, and bulk moderator temperatures, by about 5 degrees centigrade while maintaining the same margins of safety. P and L Reactors were modified for 5 psig operation in 1962, and the other 3 reactors were modified in 1963. The increased blanket gas pressure allowed about a 120-MW increase in reactor power, which was achieved in R and P Reactors in 1963 and in L, K, and C Reactors the following year.

Two new reactor fuel assemblies went into production in 1962 to increase productivity of plutonium and tritium. In February 1962, the first Mark VI-B charge began irradiation in L Reactor for production of tritium. The Mark VI-B, which had been in development by SRL since 1959, contained two concentric enriched uranium-aluminum fuel tubes sandwiched between outer and inner target tubes of lithium-aluminum. This assembly offered significant advantages over previous Mark VI type designs in temperature coefficients, productivity (grams/MWD), and cycle exposure, the latter leading to reduced component costs and higher reactor operating time (fewer scheduled outages per year). In March 1962, the first Mark V-B charge to produce plutonium was irradiated in R Reactor. The Mark V-B was an all-tubular assembly designed to replace the Mark VII-A quatrefoil design. The Mark V-B contained two concentric columns of natural uranium fuel. It was capable of higher flow and, therefore, could operate at higher power levels than the Mark VII-A, although Mark V-B charges likewise required enriched uranium “spike” assemblies. Mark V-B fuel experienced fuel swelling, however, so to combat this problem Mark V-E fuel was designed and first irradiated in 1963. Mark V-E assemblies were similar to Mark V-B except that the U-235 content was increased from that in natural uranium (0.71 wt %) to 0.95%. This increased charge reactivity and eliminated the need for spiking. It also increased both reactor power (because the slug columns were thinner and could accommodate higher flow) and productivity (as a consequence of the higher enrichment). However, the increase in productivity was achieved in tritium at the expense of reduced plutonium production, and shortly thereafter defense requirements changed in the opposite direction. A similar assembly, the Mark V-R, was therefore designed and also first irradiated in 1963. The Mark V-R was nearly identical to the Mark V-E except that the enrichment was lowered to 0.86% uranium. The lower enrichment slightly reduced total Mark V-R productivity relative to the Mark V-E, but increased the ratio of plutonium-to-tritium production.

Thus, by 1964, all the major changes had been made to the reactor fuel assemblies and to the primary and secondary reactor cooling systems to increase power level and production output. The increase in reactor power potential made possible by the various hydraulic system upgrades described above are depicted in Figure 1. In March 1967, C Reactor achieved the highest power level ever sustained in a Savannah River reactor, 2915 MW. That corresponded to more than a seven-and-one-half-fold increase over the original design power of the reactors. It

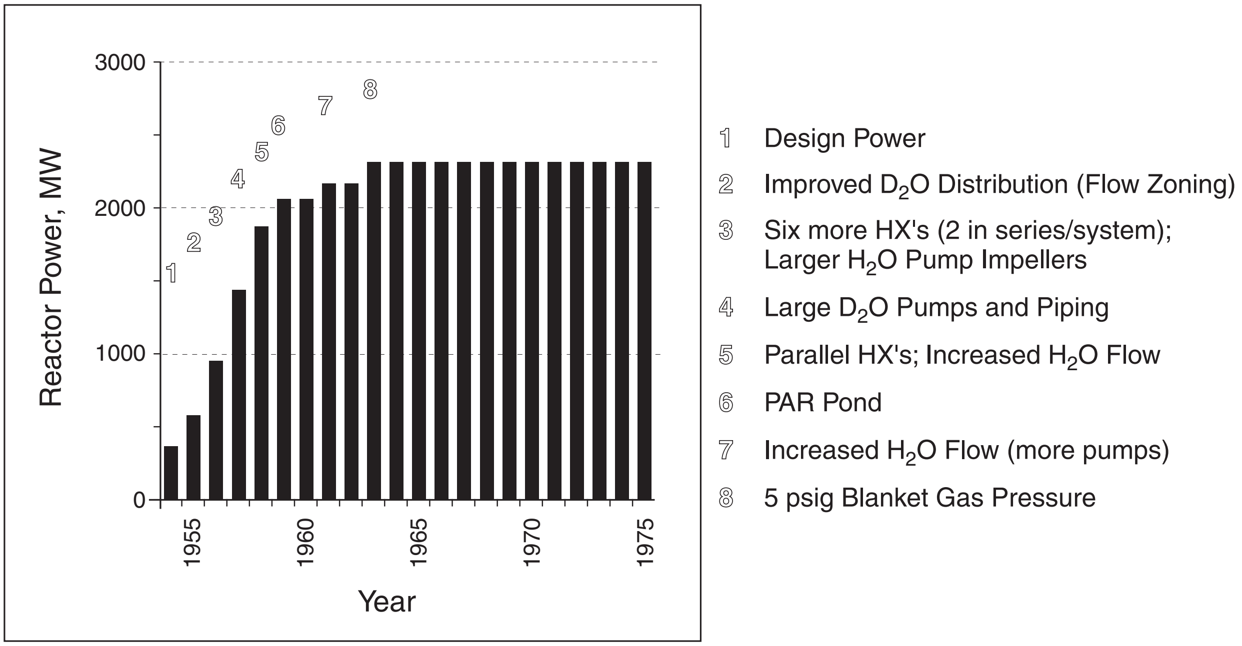

Figure 1. Nominal reactor power potential

clearly represented an outstanding achievement in the context of the contribution of the Savannah River reactors to the national defense as well as to needs in the non-defense sector.

Future work on the reactor systems beyond the mid 1960s was done to better define and improve reactor operating safety (e.g., the capability of the emergency cooling system to add light water to the reactor core in the event of a major leak from the process water system). Efforts to develop advanced fuel and target assemblies also continued. These emphasized increased productivity and versatility of designs, both for continued production of plutonium and tritium as well as for special isotopes for defense and non-defense applications (Cm-244, Cf-252, Pu-238, and others). A key development leading to more productive and versatile charges was replacing the steel semi-permanent sleeves in the upper portion of the reactors with universal sleeve housings (USHs). The aluminum USHs extended all the way to the reactor tank bottom and were the largest components that could fit through the circular holes in the reactor plenum and top shield. This facilitated design of larger diameter fuel and target assemblies. It also eliminated the time consuming and expensive effort involved in replacing the outer housings each time new fuel or target assemblies were charged to the reactors.

Taking advantage of the USH development, the ultimate tritium producer, the Mark 22 charge, began operation in 1972 and continued thereafter. The ultimate plutonium producer was a uniform charge of Mark 15 assemblies containing uranium with 1.1% uranium-235. This high enrichment resulted in a very high conversion ratio; however, it could not be accommodated in the DOE Uranium Enrichment Plants without substantial new capital investment. It was abandoned after successful irradiation of one charge was demonstrated in 1983. Instead, the Mark V-R charge for plutonium production was replaced beginning in 1968 with the Mark 14-30 charge, designed for use with the USH. Importantly, this charge utilized the “mixed lattice” concept, wherein each hexagon of assemblies surrounding a control rod cluster contained three Mark 14 driver fuel assemblies and three Mark 30 target assemblies, in alternating order. The fuel assemblies contained highly enriched uranium, and the target assemblies contained depleted uranium, leading to the term “enriched-depleted” operation. Beginning in 1973, Mark 14-30 charges were replaced by Mark 16-30 charges for production of plutonium. The Mark 16 assemblies contained more total uranium fuel than the Mark 14, leading to improved operation and economics. Mark 30 targets were gradually displaced by Mark 31 assemblies, starting in 1972, to accommodate a change in depleted uranium assay from the Uranium Enrichment Plants, from 0.14% to 0.20% U-235. Over the years, numerous variations of mixed lattice designs were used to produce special isotopes for a wide variety of defense and non-defense applications, which are beyond the scope of this paper.

Acknowledgment

The author acknowledges with sincere thanks the contribution of James M. Boswell, whose assistance in the preparation of this paper was invaluable.

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Biography

James Morrison completed his Bachelor and Masters degrees in Chemical Engineering from Syracuse University in 1954 and 1955. He then began his career as an engineer for the Du Pont Co. in the Reactor Technology Section at the Savannah River Site (SRS). He worked in various assignments at SRS and in 1967 transferred to Oak Ridge as a Du Pont representative to DOE's Combined Operations Planning group, where he helped match nuclear defense requirements to production capabilities.

Mr. Morrison subsequently joined Union Carbide Corp. and worked in managerial capacities in all three Oak Ridge plants. He was involved in a number of major projects including the Gas Centrifuge Enrichment Plant. In 1984 he rejoined strategic planning work at SRS. He served as Technical Advisor to DOE in Washington, DC in 1987-88. After returning to SRS he held key positions during the K-Reactor Restart effort and the subsequent foreign reactor spent fuel return and storage program. He retired from WSRC in 1995 and began work as an engineering consultant. Since early 1997, his major client has been the Southeastern Technology Center in Augusta, Georgia, for whom he has managed two Alternative Fuel Vehicle programs involving technology transfer from SRS.

Reactor Production Diversity

James M. Boswell

Abstract

Production diversity of Savannah River reactors was amply demonstrated by their efficiently producing of over 100 exotic radioisotopes, even though the original design mission was primarily to produce plutonium and tritium. Production versatility of Savannah River Site (SRS) reactors is unparalleled as a consequence of high power, large range of neutron flux, excellent neutron economy, and large capacity for a variety of target materials.

Operation at high neutron flux was perhaps the most outstanding advance in reactor technology made at SRS. Operation at neutron flux levels comparable to neutron flux levels of reactors specifically designed for high flux operation made SRS the principal source of synthetic nuclides for the free world from 1965 through 1970.

Records achieved during the High Flux Demonstration include a thermal neutron flux level of 6.1×10^{15} neutrons per square centimeter per second, a thermal heat flux level of 2.66×10^6 BTU per hour per square foot, and a fuel power of 226 kW per gram of uranium-235. An even higher neutron flux level of 7×10^{15} neutrons per square centimeter per second was achieved later during the Californium Production Campaign. This record neutron flux level was achieved during the time when the High Flux Isotope Reactor (HFIR) was being built at Oak Ridge, Tennessee. The peak design neutron flux level for the HFIR was $3\text{-}5 \times 10^{15}$ neutrons per square centimeter per second; a value less than that achieved in SRS reactors.

Theoretically, an SRS reactor is capable of operation at high neutron flux level while supporting up to 130 kilograms of target material compared to a maximum of 0.3 kilograms for the High Flux Isotope Reactor. However, only nine kilograms of target material was available for irradiation during the Californium-I Production Campaign.

Several world production records were achieved during High Flux Operation:

- The most californium-252 ever made (2.1 g).
- The highest specific activity cobalt-60 ever produced (700 curies per gram)
- The most polonium-210 ever made (over 0.5 kilogram)

Additional information about these and other SRS major reactor products is summarized in Table 1.

Background

Savannah River Site (SRS) reactors are believed to be the most versatile nuclear reactors ever operated. They are large, comparable in size and power level to commercial power reactors; but unlike commercial power reactors, they have been operated at neutron flux levels as high as those achieved in research reactors specifically designed for that purpose. They have proved to be extremely versatile by their ability to simultaneously produce a wide variety of radioisotopes. Over 100 different radioisotopes were produced in SRS reactors between 1954 and shutdown in 1988.

SRS reactors use heavy water (D_2O) for both coolant and moderator. This design feature contributes considerably to reactor versatility and the ability to achieve very high neutron flux levels because heavy water absorbs very few neutrons and is also an excellent media for slowing down neutrons so they can be captured in target materials.

Co-production of electricity and radioisotopes was briefly considered during conceptual design, but the idea was soon abandoned for two reasons:

- Long operating cycles are essential for efficient generation of electricity, whereas relatively short cycles are required for production of weapons-grade plutonium.
- High reactor coolant temperature is required to provide steam for turbine generators to generate electricity. High coolant temperature is incompatible with aluminum cladding. Aluminum cladding is extremely desirable because it is inexpensive, absorbs very few neutrons, and is an outstanding barrier to contain tritium in lithium-bearing targets and control rods.

SRS reactors were originally designed to operate at about 400 megawatts thermal (MWt). By 1964, the reactors were routinely operated at powers over 2400 MW_t during the winter months. In 1967, C Reactor achieved a peak power of 2915 MW_t, the highest power ever achieved in an SRS reactor. This reactor power increase of over a factor of six represents one measure of the extreme versatility that has become a hallmark of SRS reactors.

Both upgrading the reactor hydraulic system and developing sophisticated fuel and target assemblies contributed to the ability to operate at high power levels. Physical modifications required to increase reactor power are described in the article, “Increased Production,” by James M. Morrison. Sophisticated fuel and target assemblies were designed and developed at the Savannah River Laboratory to take advantage of the higher power potential of the improved reactor cooling system. Development of these assemblies is described in articles by Philip Permar and William McDonell.

In his January 1964 State of the Union message, President Lyndon Johnson ordered the shut down of several weapons production reactors due to a decreased requirement for weapons materials. As a consequence of the demonstrated versatility of SRS reactors, only R Reactor was shut down as compared to the shutdown of several Hanford reactors. This trend continued until only one Hanford reactor (N Reactor) remained on-line, while three SRS reactors continued to operate. The reason for keeping SRS reactors in service was their demonstrated capability to operate over a wide range of neutron flux levels while simultaneously producing a variety of radioisotopes.

The only reason that N Reactor continued to operate was because it co-produced about 800 MW of electricity for the Northwest. It was incapable of efficient production of both electricity and weapons-grade plutonium. Relatively short reactor cycles are required to make weapons-grade plutonium whereas long fuel cycles are necessary for efficient production of electricity. This is the very reason why co-production was ruled out during conceptual design of SRS reactors.

Reactor Description

Each Savannah River Site (SRS) reactor has approximately 600 4-inch-diameter positions that may be used for either fuel or target assemblies. Fuel assemblies normally contain U-235-bearing material while target assemblies contain the material to be transmuted into the desired radioisotope. The reactor core is the combination of all of the fuel and target assemblies. Fuel and target material can either be contained in the same assembly or in separate assemblies. A reactor core of fuel and target in the same assembly is referred to as a uniform lattice; whereas, if these materials are in separate assemblies, the core is called a mixed lattice. One of the major keys to SRS reactor versatility is the ability to simultaneously produce many different radioisotopes in a mixed-lattice core. Fuel and target assemblies are suspended vertically from an inlet process water plenum. Deuterium oxide (D_2O) is used for both coolant (dissipates fission heat) and moderator (slows down neutrons so they can be absorbed in fuel or target materials). Neutron loss due to absorption in D_20 is very low compared to other moderating materials, such as graphite or light water (H_2O). The so-called "neutron economy" is much better with D_20 moderator than with other moderating materials.

The reactor core is enclosed in a cylindrical stainless steel tank that is about 15 feet tall and 16 feet in diameter. Process water (D_20) flows down from the inlet plenum through the fuel and target assemblies to remove heat, and is then discharged into the space between the fuel and target assemblies before exiting the reactor tank. D_20 coolant exits through 6 effluent nozzles at the bottom of the reactor and is then pumped through 12 heat exchangers before return to the reactor plenum. Light water coolant flowing through the shell side of the heat exchanger dissipates fission heat to a cooling pond (R, P, and L reactors), a cooling tower (K Reactor), or a stream (C Reactor) that flows into the Savannah River.

Core Configurations

SRS reactors were originally designed and built to produce plutonium and tritium for nuclear weapons. The first reactor core consisted of four columns of aluminum-clad natural uranium slugs in each of the 600 reactor lattice positions. Reactor power was controlled by lithium-bearing control rods that produced tritium. All future reactor configurations employed lithium-bearing control rods, except those designed for high flux. Because lithium melts at a relatively low temperature, cadmium or cobalt was used in control rods for reactor cores that operated at elevated neutron flux levels, and, hence, high fuel specific power (MW_t per linear foot of fuel).

The crux of a nuclear reactor is the core design. Commercial power reactors use essentially the same basic core design for the life of the reactor. The U-235 content of fuel assemblies may vary somewhat, but the overall configuration of the core stays the same. By contrast, almost two dozen distinct reactor cores were designed, developed, and operated in Savannah River reactors. Furthermore, several additional cores were designed and developed, but were discarded for various technical reasons. Each core design is analogous to a different reactor because each design required developing fuel and target assemblies and nuclear physics experiments in test reactors to confirm the physics before irradiation in a production reactor. Each new core design required a complete safety analysis to confirm that it would perform within acceptable limits even under adverse conditions. Advanced reactor core designs were conceived and developed by engineers and metallurgists in the Savannah River Laboratory.

Almost half of the reactor cores were designed to produce plutonium for weapons. Uniform-lattice core designs consisted of either solid slugs of uranium, hollow slugs, or nested tubes. Hollow slug and nested tube designs were developed to increase both flow area and heat transfer surface area. All of the slug designs used natural uranium (0.71% U-235). Nested tubular designs ranged from natural uranium to 1.1 wt % U-235. More flow and heat transfer area contributed to higher reactor power while higher U-235 content contributed to increased amount of Pu-239 produced per megawatt day.

High Flux Demonstration

The crowning technical achievement of SRS reactors was operation at high thermal neutron flux in C and K Reactors during the mid 1960s. In 1961, President John F. Kennedy appointed Dr. Glenn T. Seaborg as chairman of the Atomic Energy Commission. Prior to that appointment, Dr. Seaborg shared the 1951 Nobel Prize for chemistry with Edwin M. McMillan for the discovery of artificial elements heavier than uranium, including plutonium, americium, curium, berkelium, and californium. Naturally, Dr. Seaborg had a strong interest in production of sufficient quantities of these artificial elements for scientific evaluation, which might lead to a commercial market once their properties were fully known. Hence, Dr. Seaborg was instrumental in directing the Savannah River Operations Office to authorize high flux operation in a Savannah River reactor to produce gram quantities of californium and other transuranium isotopes.

In the same time frame, a high flux isotope reactor (HFIR) was designed and built at Oak Ridge to produce small quantities of artificial nuclides for scientific evaluation. Operation of an SRS reactor before completion of the HFIR would provide an opportunity to produce target material (plutonium, americium, and curium) for the HFIR and also to evaluate the reactor performance of target assemblies that would later be used in the HFIR.

Radioisotopes that require multiple neutron captures and that have short half-lives can only be made in quantity by irradiating target material at high neutron flux levels. Neutron flux level is directly proportional to fuel specific power (megawatts per foot of fuel assembly) and inversely proportional to fuel U-235 content (grams of U-235 per foot of fuel assembly). The most efficient production of plutonium and tritium is achieved at a thermal neutron flux level of about $7 - 10 \times 10^{13}$ neutrons per square centimeter per second. By contrast, optimum production of Cf-252 requires a hundredfold higher neutron flux level because of the multiple neutron captures required to produce it, and the fact that its half-life is only about 2.5 years.

A decision was made to demonstrate high flux operation in C Reactor prior to the actual production campaign in K Reactor. To achieve high flux, the number of fuel positions was decreased from approximately 600 to 107, the active length of U-235 in the fuel assemblies was reduced from about 12 to 6 feet, and the fuel loading was reduced to 25 grams of U-235 per foot. This combination of changes resulted in increasing the neutron flux level from about 7×10^{13} to 6×10^{15} neutrons per square centimeter per second.

Target material consisted primarily of Pu-242. In addition, three 1-inch-diameter thimbles contained Am-243, Cu-244, and 150 nuclides of 66 elements for 9 universities and laboratories.

Control rods for the high flux demonstration contained cadmium, which could withstand operation at higher temperature without melting the lithium, which is normally used to produce tritium in low flux operation. Cadmium in control rods was replaced with cobalt in the following Californium Production Campaign.

The High Flux Demonstration began in February 1965 and lasted a year. A neutron flux level of 6×10^{15} neutrons per square centimeter per second was achieved, and the demonstration was considered a success by paving the way for the following Californium Production Campaign.

Production of Radioisotopes

In addition to the relatively large-scale production of radioisotopes described in the following sections, small amounts of 150 different target materials were irradiated primarily during high flux operation. Irradiation of these samples was requested for research by Argonne National Laboratory, Oak Ridge National Laboratory, and the University of California laboratories at Berkeley and Livermore. A list of samples irradiated during the High Flux Demonstration is contained in DP-999, “The Savannah River High Flux Demonstration.”

Californium-252

Based on technical information gained through operation of the High Flux Demonstration in C Reactor, an improved reactor core was developed to produce Cf-252 in K Reactor, which began operation in August 1969. The improved core consisted of 96 large-diameter assemblies, each of which contained 3 concentric tubes of U-235 alloy fuel rather than the 2-tube assemblies used for the High Flux Demonstration. Eighty-six of the fuel assemblies contained Pu-242 in a four-foot section of the outer housings. Irradiated fuel was discharged separately from the outer housing, which remained in the reactor until termination of the production campaign. The reactor core also contained six assemblies of Am-243 and Cm-244 target material. Each fuel cycle lasted 3.5 to 4 days, and was followed by a 12 to 18 hour reactor shutdown to replace irradiated fuel tubes.

The reactor core of fuel assemblies was surrounded by 104 target assemblies containing Np-237 to produce very pure Pu-238. These assemblies were located in the reflector between the core and the reactor tank wall.

A peak flux of 7 x 10^{15} was achieved, and 2.1 grams of Cf-252 were produced before termination of the production campaign on November 9, 1970, due to failure of an antimony-beryllium source rod, while the reactor was being refueled. A second Californium Production Campaign was planned, but it was not executed because the market for Cf-252 never materialized as originally expected.

In 1965, Carl Schlea and Dean Stoddard of the Savannah River Laboratory proposed using Cf-252 in needles for cancer therapy. The key to this proposal is the idea of treating localized cancers with neutrons from Cf-252 encapsulated in needles similar to those used to contain radium. The problem with radium needles inserted in a tumor is that radium emits X-rays, which do not discriminate between cancerous cells and healthy cells; hence, treatment can be taken only with the risk of destroying healthy tissue. Basing their research on theories previously developed, Schlea and Stoddard concluded that Cf-252 might provide a lethal dose of radiation to a tumor without irreparable damage to adjacent normal cells. They recognized that treatment would be most effective for tumors located too close to vital organs to be removed surgically.

As a consequence of the Schlea-Stoddard proposal, a Cf-252 needle manufacturing facility was designed and installed in the Savannah River Laboratory. Over 3000 needles were made that contained from 5 to 140 micrograms of Cf-252 for evaluation by numerous hospitals and universities here and abroad.

Tritium

The Savannah River Plant was built primarily to produce tritium, an essential ingredient in thermal-nuclear weapons. Tritium significantly increases the yield of nuclear weapons. Over the operating life of the reactors, six different reactor cores were designed, developed, and implemented to produce tritium. All of the tritium-producing cores were of uniform-lattice design. That is, both the fuel (U-235) and the target material (lithium) were contained in the same assembly, but in separate slugs or tubes within the assembly.

The Mark 22 was the most efficient of the tritium-producing cores because it was designed to use the full capacity of the reactor hydraulic system and was compatible with the use of outer aluminum housings that could be recycled for several years of operation. In previous designs, the outer housing contained lithium and was discharged from the reactor with the rest of the assembly each time the core attained its goal exposure (megawatt days).

One tritium-producing core (Mark VI-E) was designed with a removable inner lithium target so that the fuel could be irradiated to a very high exposure. The high exposure depletes about 70% of the U-235 and thus concentrates U-236, which is formed from every sixth neutron capture in U-235. Irradiation of U-236 forms neptunium-237 (Np-237), which is the target material for production of Pu-238. Hundreds of kilograms of tritium were produced during the 34-year operating life of the SRS reactors.

Plutonium-239, -240, and -242

Plutonium for weapons was produced in one or more SRS reactors every year from initial operation in 1954 through the final shutdown in 1988. Approximately half of the two dozen reactor cores that were operated in SRS reactors were designed primarily to produce plutonium. The initial cores contained the fissile isotope (U-235) and the target isotope (U-238) in the same assemblies (uniform lattice). Mixed-lattice reactor cores to produce plutonium began operation in 1968. The mixed-lattice core consisted of an equal number of highly enriched uranium (U-235) fuel assemblies and depleted uranium (U-238) target assemblies. The fuel assemblies supplied the neutrons, and the product (Pu-239) was produced in the target assemblies.

The most productive plutonium-producing reactor core (Mark 15) was a uniform lattice of slightly enriched uranium (1.1 wt % U-235) in assemblies of concentric tubular slugs. The outermost 84 reactor positions contained lithium-bearing tubular assemblies, which protected the reactor tank wall from excessive neutron damage and reduced argon formation between the reactor tank and the surrounding thermal shield.

Plutonium was produced at assays that ranged from 3% to about 40% Pu-240. Weapons-grade plutonium is defined by both the Pu-240 and Pu-241 assays. The very high assay Pu-240 material was produced for use as target material for production of higher radioisotopes and for breeder reactor studies that use plutonium as fuel. Hundreds of kilograms of Pu-239 and Pu-240 were produced between 1958 and 1984.

Tens of kilograms of Pu-242 were produced by irradiating large quantities of Pu-239 over a period of several years.

Plutonium-238

Plutonium-238 generates 0.5 watt per gram when it decays by spontaneous emission of alpha particles, which are easily blocked by a thin metal sheath. These properties coupled with a relatively long half-life (about 89 years) make Pu-238 an ideal heat source for use in thermoelectric generators for space exploration. SRS began production of Pu-238 by the irradiation of Np-237, a byproduct of the fission of U-235. Experiments left on the moon by "Apollo" crews were powered by Pu-238 made at SRS. Unlike solar cells, generators powered by Pu-238 are ideal for exploration of the outer planets.

The challenge in making Pu-238 is to minimize formation of Pu-236, which is formed by absorption of fast neutrons. The solution to this problem was design of reactor lattice configurations that maintain adequate separation of fast neutrons from neptunium target material. To accomplish this separation, neptunium targets were located in the D_2O reflector beyond the fuel in high flux cores.

By 1978, over 300 kilograms of Pu-238 had been made in SRS reactors.

Another route to formation of Pu-238 that avoids the offensive Pu-236 isotope is producing it as a decay product of Cm-244. In 1970, Pu-238 was separated containing less than 0.3 parts per million of Pu-236

Curium-244

Curium-244 is produced through successive neutron captures in Pu-239. Curium -244, like Pu-238, decays by alpha emission but produces five times as much heat (2.5 watts per gram) as Pu-238. However, the half-life of Cm-244 is only about 18 years compared to about 89 years for Pu-238; hence, Pu-238 is the preferred isotope for long-term space missions.

Gram quantities of Cm-244 were first produced in SRS reactors in 1962. In 1964, a special reactor core began irradiation to produce kilogram quantities of Cm-244 for evaluation as an alternative to Pu-238 and for use as target material to produce Cf-252. The reactor core for this production campaign (called Curium I) was designed to operate at a higher neutron flux level than previous SRS reactor cores. The targets from this irradiation were reprocessed and made into new targets for the second phase of this program (called Curium II). By 1967, a total of 5.9 kilograms of Cm-244 had been made during the two curium production campaigns.

Continued irradiation of plutonium target material eventually produced about 12 kilograms of Cm-244.

Uranium-233

A breeder reactor produces more fissionable material than it consumes to maintain a nuclear chain reaction. Uranium-233 (U-233) is such a material when used in conjunction with thorium targets. Uranium-233 is produced by neutron capture in thorium, which is an abundant element in nature.

Admiral Hyman G. Rickover was a key figure in the development of the U.S. nuclear navy and commercial reactors. One of his projects included developing a breeder reactor using the Shippingport Reactor located in Pennsylvania. SRS was requested to produce several hundred kilograms of U-233 for Admiral Rickover's program.

By 1956, U-233 was being produced in small quantities, but it was contaminated with U-232, which emits strong gamma radiation and, hence, made fuel fabrication from this material very difficult. In the original irradiations, the fissionable U-235 and the thorium targets were in the same assemblies. Since U-232 is produced from interaction with fast neutrons, the challenge was to separate the fissionable U-235 from the thorium target material. This separation of fuel and target material allowed the neutrons reaching the thorium to be slowed down by the D_2O moderator, thus greatly reducing the formation of U-232.

By 1965, U-233 was being produced in quantity with only 3 - 6 parts per million of U-232. Several hundred kilograms of U-233 were produced through 1968.

Polonium-210

Polonium-210 is made by irradiating targets of bismuth-209. Operation at high neutron flux with small reactor cores during the 1960s provided an opportunity to demonstrate the production of large quantities of Po-210 by irradiating bismuth targets in the D_2O reflector between the reactor core and tank wall. Approximately 0.6 kilogram of Po-210 was made that way.

Use of Po-210 in thermoelectric generators was briefly considered, but this application was abandoned because of its short half-life (130 days) and difficulty in separating polonium from bismuth.

Cobalt-60

Irradiation of Co-59 produces Co-60, which emits very penetrating gamma radiation. Sources made of Co-60 can be used for cancer treatment, food sterilization, radiography, and as heat sources for thermoelectric generators.

Small amounts of low-specific-activity Co-60 (~ 50 curies per gram) were produced in SRS reactors as early as 1955. High flux operation, beginning in 1965, provided an opportunity to make large quantities of very-high-specific-activity Co-60 because Co-59 was the preferred material for control and safety rods in high flux reactor cores. Lithium, normally used in control rods, melts at a considerably lower temperature than cobalt. Therefore, for reactor safety reasons, Co-59 was substituted for lithium during high flux operation, where a Co-60 specific activity of 700 curies per gram was achieved (the highest specific activity Co-60 ever made). Specific activity of pure Co-60 is about 1140 curies per gram.

Perhaps the greatest interest in Co-60 was manifested by the food industry, which used gamma radiation to sterilize food for long-term storage without refrigeration.

Table 1. SRS Reactor Major Products

Radioisotope	When Produced	Amount	Application
Plutonium-239	1954 - 1988	1000's kg	Nuclear Weapons
Tritium	1954 - 1988	100's kg	Nuclear Weapons
Uranium-233	1956 - 1968	100's kg	Breeder Reactor Development
Plutonium-238	1959 - 1988	100's kg	Thermoelectric Generators for Space Exploration
Plutonium-240	1958 - 1984	100's kg	Target Material for Transplutonium Isotopes
Plutonium-242	1964 - 1984	10's kg	
Cobalt-60	1956 - 1970	~ 66 mega curies	Gamma Radiation Source Heat Generation Sources
Curium-244	1962 - 1978	~ 12 kg	Thermal Electric Generators Target for Production of Transplutonium Isotopes
Polonium-210	1966 - 1969	~ 600 g	Intense Radiation Source
Californium-252	1965 - 1970	2.1 g	Cancer Treatment, Oil-Well Logging, etc.

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Definitions

Neutron Flux - Neutrons per square centimeter per second

Fuel Specific Power - Megawatts of thermal heat output per linear foot of fuel

Half-life - The time required for half of a radioisotope to disappear through radioactive decay

Target Material - Material to be irradiated with neutrons for transmutation to different radioisotopes

Pure Plutonium-238 - Contains less than 0.3 ppm of plutonium-236, which is a contaminant due to its gamma radiation

SRS - Savannah River Site.

MW_t - Megawatt thermal (heat produced by nuclear fission)

Co-production - The simultaneous production of nuclear materials and generation of electricity

Uniform Lattice - A reactor core in which all of the assemblies are alike

Mixed Lattice - A reactor core containing a mixture of uranium-235-bearing assemblies and target assemblies

Neutron Economy - Fraction of useful neutrons

Biography

James Milton Boswell

(B. S., Mechanical Engineering, University of Louisville)

1953 - Savannah River Plant, Works Technical Department; Engineer, Heavy Water Technology; Engineer, Reactor Technology

1959 - Process Supervisor, Reactor Technology

1962 - Senior Supervisor, Reactor Technology

1963 - Savannah River Laboratory; Nuclear Material Division; Reactor Engineering Division

1965 - Research Supervisor, Advanced Planning

1968 - Advanced Operational Planning

1969 - Research Manager, Reactor Engineering Division

1974 - Research Manager, Advanced Operational Planning

1977 - Director, Nuclear Reactor Technology Section

1981 - Special Assignment, Technical Advisor to the Director of The Office of Nuclear Materials Production; U.S. Department of Energy, Germantown, MD

1982 - Director, Nuclear Materials Production Support Group

1986-89 - Principal Consultant, AED Planning

1989-99 - Private Consultant, President of J. M Boswell, Inc.

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The Restart of L Reactor

Thomas C. Gorrell

Abstract

L Reactor was returned to a fully operational condition, incorporating safety improvements and other modifications already in place in the three operating reactors. Restart criticality was achieved in October 1985. The reactor had been in a standby status from 1968 until 1980. Substantial repair, renovation, and new capital installation took place during the five-year restart effort. The peak work force was approximately 240 operations and 800 construction personnel. At the time, the project was the largest single Savannah River Plant (SRP) construction project since the original plant startup. This paper describes some of the problems and highlights of the unprecedented restart achievement.

Introduction

The reactor console operator made a small adjustment in control rod position, reviewed his nuclear instrumentation, then announced, “We are critical at low power.” This quiet understatement symbolically marked the climax of an ambitious five-year effort to rescue the L-Reactor complex from a standby condition and restore it to full operational capability. This paper describes a few of the problems and highlights of that achievement, which culminated with restart criticality on October 31, 1985.

L Reactor was the third in the sequence of the five Savannah River Plant (SRP) production reactors to be brought on line during the original SRP startup. It began operating in mid 1954 and was operated until February 1968, when a reduced need for nuclear material for the U.S. defense program led to its retirement from service. It had been in a standby status for more than 12 years when the restart effort began in October 1980.

Need for Product

The Cold War and nuclear arms race between the United States and the Soviet Union were still in full swing in the late 1970s. Nuclear Weapons Stockpile memoranda prepared at that time showed that the U.S. production rate of weapons material was not sufficient to meet projected needs of the Defense Department. Some of the options that were considered to increase production included developing a new fuel assembly for the still-operating P, K, and C Reactors at SRP, restarting a Hanford reactor, recovering plutonium from commercial reactor spent fuel, and restarting L Reactor at SRP. The Department of Energy (DOE) used a review process consistent with the National Environmental Policy Act (NEPA), and ultimately selected L-Reactor restart as the preferred choice. Although the goal startup date was set as October 1983, there was no precedent for restarting a reactor after a 12-year shutdown. Funding of approximately $200 million was authorized for the entire project.

Ironically, it was President Jimmy Carter who authorized the program that ultimately led to L-Reactor restart. Only three years earlier (1977), President Carter had issued an executive order that prohibited reprocessing spent fuel from commercial nuclear power reactors, including plutonium recovery. President Carter compartmentalized these two executive actions as part of two completely separate technical and political issues.

Condition of L Area

Several sources describe L Area's official status following its 1968 shutdown as "standby", which Webster defines as "something that can be relied upon". In fact, L Area was not in good condition. After operating personnel completed their shutdown program in 1968, no significant maintenance was performed on any system. Equipment inside the main process building (Building 105-L) was protected from the sun and rain, but no temperature or humidity control was provided. However, most major process systems were intact because parts cannibalization had been discouraged during the standby period.

The initial group of restart personnel that arrived at the area on October 14, 1980, found primitive conditions. Outside support buildings and their contents had deteriorated, especially equipment made of carbon steel. The area was overgrown with weeds, grass, and small trees. Animals and birds occupied some of the buildings, including a family of bobcats in a maintenance shop and flocks of pigeons around Building 105-L. The first action the team found necessary was to set up electrical power, ventilation, and sanitary facilities (exterior portalets) for Building 704-L, simply to make the area habitable. These humble beginnings made it apparent to everyone that a long, difficult journey lay ahead.

Initial Major Issues

Several major issues had to be addressed immediately:

- What were the sources of manpower available to staff the project? Only a limited number of people could be transferred from the operating reactors without adversely impacting their operation. In fact, the project ultimately was staffed by a combination of transfers of a few experienced people from several plant departments, attrition from other plant areas, and new hires.
- What was the physical condition of the equipment? A few cursory inspections had been conducted to assure that restart was practical, but more detailed inspections were needed, and soon. Early decisions had to be made about what equipment could be repaired and what must be replaced.
- What management approach should be used to assure that all systems were accounted for and made operational on a practical, organized schedule? Key systems had to be identified to receive priority attention.

The Beginnings

Staffing was begun at once with an initial group of 12 handpicked people, all volunteers. The operations staff size ultimately peaked at about 240 people in 1983. Management of the restart team stressed from the outset that the project was unique and that everyone on the team had the opportunity to make a special contribution towards its success. New hires, in particular, could learn about SRP reactors as they took part in inspections and interacted with experienced staff members. Several of the new hires were female engineers, reflecting the national trend of increased involvement of females in the professional, technical workforce.

The group was named the "L Startup Project Team" (LSPT) with emphasis on "team". The LSPT mission from Day 1 was

- on time
- under budget
- no injuries

The project logo was a phoenix, a mythological bird consumed by fire only to be reborn and spring up again from its own ashes. The logo adorned the cover page of most documents prepared by LSPT personnel and was painted on a huge sign hung conspicuously from the highest elevation of the reactor building. A concerted effort was made to maintain a high level of morale throughout the restart effort.

The LSPT organization was arranged into four major groups, Design Liaison, Operations, Works Engineering, and Design & Technical. The function of these groups was much like that of SRP departments having similar names. However, under the Project Management Team (PMT) system pioneered by Du Pont at Victoria, Texas, all LSPT personnel reported to a single superintendent whose office was in L Area. This arrangement, a new concept for SRP, proved to be effective in the decision-making process and contributed significantly to the ultimate success of the project. Engineering Department Design and Construction personnel operated under a separate chain of command.

Early decisions were made to define major areas of responsibility. Most design and construction work was to be done by the Du Pont Engineering Department, based on requirements and specifications provided by LSPT. Also, LSPT would provide for liaison among the other groups as well as provide overall technical support. For funding accountability, restart work was divided into “capital” for new equipment and “cost” for repair or renovation of existing equipment.

The management approach used to attack the project was to divide the complex into 28 design areas and assign responsibility for each design area to one or more engineers. Several documents would be prepared for each design area. Each document would have a specific purpose and objective, and the full set would comprise a consistent approach in achieving the restart of all systems. The documents included:

- Work Scope − a short description of the work required to restore the system to service.
- Basic Data Report − a technical description of the system. (The basic data report concept was not invented for the restart of L Reactor. It was widely used by Du Pont to document the requirements for new equipment or systems).
- Quality Assurance Assessment − an evaluation of critical parts of the system that would require special attention and control in implementing the new SRP QA program.
- Quality Assurance Action Plan − the vehicle for ensuring that the specifications were met.
- Job Plan − a detailed procedure that ensured a system was in a safe condition to be worked on and was returned to a safe operable condition after work was completed.

The preparation of these documents often served as a learning process, as the engineer found it necessary to research, review, and fully understand his/her system before publishing the report. Preliminary scopes of work for each design area and a schedule were provided to DOE and the Du Pont Engineering Department by December 1980. The project was underway.

Equipment Inspections

Detailed equipment inspections were begun as soon as it was safe for personnel to enter the process areas. The Equipment Engineering Section of SRP conducted many of the inspections. The results were needed early to determine which systems would require the most attention. Several techniques were used, including visual, dye penetrant measurements, ultrasonic thickness measurements, and eddy current testing. In many cases, specialized apparatus was built to conduct a satisfactory examination.

No major surprises or disappointments arose from the inspections. In fact, much of the equipment was in better condition than expected considering the lack of maintenance. About 30 specific recommendations were made for repair, replacement, or overhaul of equipment, including the following examples:

- Purchase new heat exchangers.
- Replace all carbon steel cooling water pipes having diameters of 6 inches or less.
- Remove deposits of aluminum nitrate from the reactor vessel and effluent nozzles.
- Remove all asbestos insulation from the area.

The LSPT Effort

The preparation of Basic Data reports and QA Assessments began early in 1981. These reports were reviewed, revised, and approved for release. The approved versions of Basic Data reports were transmitted to the Engineering Department late in 1981. QA Action Plans were ultimately issued by the Engineering Department to specify requirements during design, procurement, and construction of new capital equipment.

Planning and scheduling were heavily emphasized throughout the restart effort using computer programs, graphs, charts, and countless dreary planning meetings. Critical-path schedules and work-accomplished charts were updated daily. Some difficulty was experienced in developing work schedules that were mutually agreed upon by LSPT and the Engineering Department.

Each design area has its own history of document preparation, inspection, restoration, and testing. It would be impossible to describe such details in this brief paper. As a substitute for this detail, the reader might simply try to visualize about 240 operations people and 800 construction people at one site, hard at work for almost 3 years in a construction zone. An enormous amount of reports and documents was generated. As expected, there were some delays, some milestones not met, and some disappointments, but there was never any serious doubt about the final outcome. The objective was to bring all these systems together, ready for safe, reliable operation.

Renovation of most systems was complete by late 1983 but environmental issues would delay nuclear operation until 1985. Operational tests were not complete. Management decided to assimilate the respective divisions of LSPT into existing SRP departments. The LSPT organization formally ceased to exist on September 1, 1983. Responsibility for outstanding punch-list items and final systems testing was transferred to the respective SRP departments. Cost accounting records show that $186 million had been spent on the project, compared to the $214 million authorized.

A highly complimentary letter of appreciation was written by Richard Denise, acting DOE manager at SRP, to Gerald Curtin, vice president of Du Pont Petrochemicals Department. The letter commended all those who contributed to the success and safety of the project, recognizing that it was the largest single construction effort at SRP since the original startup in the early 1950s. Members of the LSPT team still feel a sense of pride to have been part of the project.

Environmental Issues

An enormous amount of time and energy was expended to address the many environmental issues that were raised about the restart of L Reactor. The final resolution of these issues had far-reaching implications and would later prove to have adverse effects on reactor operation. One major point of contention involved "thermal mitigation," or reducing the environmental effects of the heated cooling water as it left the area.

In 1982, DOE published an environmental assessment on the proposed restart of L Reactor, with a finding of No Significant Impact. Subsequently, several environmental concerns were raised by groups such as the Natural Resources Defense Council (NRDC) and the Attorney General's office in South Carolina. A lawsuit was filed in November 1982 to require the preparation of a detailed environmental impact statement (EIS) before startup. DOE committed to preparing the EIS, and a Notice of Intent was filed in July 1983.

The EIS process ran the full gamut of document preparation, hearings, and solicitation for comments before the extent of system modifications required for compliance was known. A significant requirement was the construction of a 1000-acre lake near the L-Reactor outfall to receive the cooling water discharge. The reactor was to be operated in such a way that a temperature of 90 degrees F or less was maintained in about half of the lake. Allegedly, this condition would contribute toward establishing a balanced biological community in and around the lake three to five years after operation was resumed.

This extensive involvement of federal and state agencies was unprecedented in SRP history. These included the federal Environmental Protection Agency (EPA) and the South Carolina Department of Health and Environmental Control (SCDHEC). The process that evolved during the L-Reactor restart project would impact strongly on the way these same agencies would conduct similar business in the future. The point of regulation had been moved from the plant boundary and the Savannah River to areas within the SRP site itself.

Special Capital Systems

Several new systems were installed in P, K, and C Areas during the L-Reactor standby period and had to be built especially for L Area. These systems included:

- The M-2 console − a hard-wired logic system to determine the need for light water addition to the fuel in the event of a severe process water leak.
- Computer-based systems for control rod operations, flow and temperature monitoring, and selective fuel charge/discharge operations.
- Diagnosis of Multiple Alarms (DMA) − a computer-based system that assisted control room operators in interpreting and prioritizing alarm messages before taking corrective action during emergency situations.
- Improved emergency cooling system and water removal system.

In addition, the procurement of the 12 process water/cooling water heat exchangers deserves comment. Examinations of the original L-Area and R-Area heat exchangers revealed that many were in poor condition, and repair would not be cost effective. Consequently, bids were let for the purchase of new units. After no bids were received from U.S. firms to build the shells and heads, contracts were awarded to Mitsui Engineering and Hitachi Engineering in Japan. One might not expect that a country in which two cities had been devastated by U.S. nuclear weapons 35 years earlier would choose to be part of the U.S. nuclear weapons program, even for a profit. The new heat exchangers were delivered on time and operated satisfactorily. Their design incorporated improvements to reduce the probability of tube failure and leakage.

Problem Areas

Two examples of systems where unusual problems were encountered were the fuel assembly charge/discharge (C&D) machines and the disassembly area basin.

- The C&D machines and associated hardware were highly complex devices used for remote transfer of new fuel assemblies and other components <u>to</u> the reactor, and transfer of irradiated assemblies <u>away</u> from the reactor to underwater storage. The machines in P, K, and C Areas had been modified several times by separate projects during the 12-year L-Reactor outage, but there were no as-built drawings in October 1980. The only workable approach for the L-Reactor restart was to modify the machines sequentially for each project and accept the inefficiency that occurred.
- Renovation of the disassembly basins was also a challenge. (The basins were used to store irradiated materials after discharge). They had been kept full of water during the L-Reactor standby period. A thick layer of sludge covered the floor bottom, with several miscellaneous radioactive components buried in it. As the water was drained and the walls dried, care had to be exercised that contaminants did not become airborne. Also, adequate shielding had to be maintained for irradiated materials on the floor. After many months of tedious, careful cleanup, the basins were finally emptied, scraped, and repainted.

An additional, chronic problem that proved troublesome in several systems was availability of correct as-built drawings and schematics specific to L Area. The system engineers often found it necessary to verify or update the prints by field inspection.

Health Protection and Safety

The potential for unusual health protection or safety incidents was high in L Area during restart. Health Protection and Safety programs were implemented to minimize radiation exposure and prevent injury. When water or instrument lines were opened for the first time, personnel were required to wear plastic suits supplied with breathing air. Similar care was taken as each new system was inspected. As a result, the number of tritium uptake and skin contamination cases was kept at an acceptably low level. A fire brigade and an emergency rescue team were established.

Some examples of unique jobs are:

- Welding or cutting cooling water lines that might contain methane, an explosive gas
- The odious task of removing 12 years of pigeon fecal waste from the stack area
- The high radiation environment created during the removal of irradiated sleeves and plugs from the reactor tank top
- The extensive use of special paint containing xylene
- Removing asbestos from several systems.

Some unusual safety-related situations did arise, often related to the co-occupancy of facilities by LSPT and Construction personnel. Both groups had full-time safety engineers assigned to the project. From January 1981 through September 1983, no operations employee sustained a Restricted Workday or Lost Workday class injury. Construction sustained two Lost Workday Cases early in the restart, then remarkably accumulated 2.5 million injury-free exposure hours through September 1983. The safety performance during the restart effort was outstanding.

Contingency Tests

The restart schedule was revised drastically after the requirement to prepare a full EIS was imposed. To exploit the availability of this time, several special "contingency tests" were planned and conducted. The intent of the tests was to learn more about the behavior of key systems during abnormal conditions. The test results would be applicable to all SRP reactors. Examples of the tests are:

- Confinement Heat Removal (CHR) − several below-grade rooms were intentionally flooded with about two feet of water using the emergency addition systems. The test objective was to demonstrate that emergency coolant could be supplied and removed by the existing systems. The test was successful.
- Plenum Level/Gradient − hydraulic pressures were measured in the primary system at reduced flow conditions. The data were used to calculate the flow available to fuel assemblies during emergency situations when the coolant flow might be very low.
- Ventilation − airflow data were taken in the air plenums at the filter compartments to assist in optimizing the removal of contaminants from the reactor room air.

Final Preparations

Slowly, finally, all parts of the project began to come together. The copier machine ran constantly and the document storage room overflowed with reports. Operations groups prepared and conducted functional tests of all the systems to demonstrate operational readiness. Extensive training sessions were held for reactor operators and supervisors. QA work was completed, audited, and approved. DOE teams conducted operational readiness reviews. A startup assessment committee consisting of over 50 SRP and Savannah River Laboratory engineers reviewed the status of all systems to assure satisfactory completion of all outstanding work items. The Management Oversight Committee issued an approval report, contingent on the successful completion of a few tests.

By late October 1985, all the formal authorizations necessary for nuclear startup were finally in place.

Reactor nuclear startup was an anticlimax compared to the rush of activity that preceded it. With few dignitaries in attendance, low power criticality was achieved at 4:42 p.m., October 31, 1985. All the reports, meetings, tests, and hard work were finally rewarded by a successful restart. As fate would have it, the new Phoenix was born on a Halloween night.

Epilogue

An ideal follow-up to a story about the successful restart of L Reactor would be to report that the reactor was operated for several years at the expected powers of 2000 MW or greater. However, that is not the case. During winter operation, L Reactor did achieve power levels as high as 2700 MW, limited by conventional thermal/hydraulic limits. However, during hot, summertime operation, the L-Lake temperature limit of 90 degrees F often restricted power to a few hundred megawatts. This extreme, seasonally dependent variation in power became the routine form of operation until the final shutdown in June 1988. Fortunately, the achievement of successfully restoring L Reactor to operational readiness is undiminished by its final operating history.

Biography

Thomas Gorrell received a BS degree in physics from Penn State in June 1953, and was part of the initial startup activity at SRP. His first work assignment was with the Process Development Pile (PDP). In October 1953, the PDP was made critical with a Mark I fuel charge replicating the first R-Reactor charge. (R Reactor was the first of the five [R,P,L,K, and C] reactors to be operated, beginning in December 1953.)

Following a tour of duty with Reactor Technology in R, C, and L Areas, Mr. Gorrell joined the physics group of the Heavy Water Components Test Reactor.

In 1981, he joined the LSPT group and took part in the initial restart activities.

Mr. Gorrell's final work assignment in 1988-1989 was to act as liaison between Du Pont management and the incoming Westinghouse management who conducted an independent safety review of the SRP reactors. He retired as a research associate in March 1989, when Du Pont completed its contractual obligations at SRP.

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JOSHUA—A Nuclear Reactor Design and Analysis Computational System

John W. Stewart

Abstract

JOSHUA is a computational system developed to perform the extensive engineering and physics calculations needed in the design and analysis of Savannah River Plant (SRP) nuclear reactors. Application of this system significantly enhanced the efficiency, effectiveness, accuracy, and safety of nuclear reactor design and analysis. The unique features of SRP reactors, which necessitated the Site's development of its own, unique design and analysis capability, are described. Short histories of computer science and reactor physics and engineering, and the state of those sciences when JOSHUA development began in 1968, are presented. The first transistor-based computers, with random-access disk storage and computer terminals, had just become commercially available. SRP had used the technical developments from the Manhattan Project and had developed extensive experimental data describing SRP reactor phenomena. The JOSHUA system, including both the operating system and the applications system, is described. The importance of such a modular, data-based computational system for the multi-step, iterative reactor calculations is explained. The significance of this development is discussed in terms of its impact on SRP reactor design and analysis, its impact on the development of integrated computational system for reactor design and analysis, and its impact on computer science.

Introduction

JOSHUA[1] is a computational system developed to perform the extensive engineering and physics calculations needed in the design and analysis of Savannah River Site (SRS) nuclear reactors (Honek 1975). Development of the system began in 1968 and continued for about a decade. JOSHUA is a modular, data-based scientific computing system. It comprised an operating system to facilitate data management, program execution, computer terminal use, and an applications system to perform the numerical calculations representing the science and engineering models. JOSHUA applications represent an improved organization and enhancement of science and engineering models based upon the powerful theoretical, mathematical, and experimental methods that were developed during the Manhattan Project and improved in subsequent years. The JOSHUA Operating System was revolutionary in that it used new computing technologies that were just beginning to become commercially available. These technologies included solid-state electronics for both memory and digital computation, random access data storage disks, and computer terminals to display alphanumeric characters and graphics. At Savannah River, safety considerations were always paramount in the design and operation of the nuclear reactors, and JOSHUA applications reflect this priority. The use of the JOSHUA system significantly enhanced the efficiency, effectiveness, accuracy, and safety of nuclear reactor design and analysis.

Development of JOSHUA in Historical Context

The Savannah River Plant Reactors

The Savannah River Plant, now Savannah River Site, was built to produce the nuclear materials, principally tritium and Pu-239, required by the United States nuclear weapons program. When the United States government announced its decision to build the Site in 1950, there were very few reactors in the world, almost all in the United States, and each of these reactors was unique. By the late 1950s, a number of companies in the U.S. and other countries were developing the reactors that would form the basis of the nuclear power industry for commercial electrical generation. These commercial ventures focused on relatively few reactor types and sought the economic benefits of reactor standardization. Because the Savannah River Plant reactors were intended for materials production, they were of a very different design than the nuclear reactors that were built later to produce electricity and for marine propulsion. The SRP design was strongly influenced by experience in design and operation of the production reactors built at Hanford, Washington, during the Manhattan Project. Some of the characteristics that distinguished Savannah River reactors from other reactors, especially power reactors, are the following:

- Low pressure and low temperature operation
- High specific power (i.e., heat generation rate per unit volume produced in fuel materials)
- Very high heat fluxes (i.e., heat transfer from fuel materials to coolant)
- Heavy water (i.e., D_2O) moderator and coolant
- Downflow of coolant through fuel assemblies
- Metal fuel materials, generally aluminum alloys clad in aluminum, rather than ceramic fuel materials
- Calandria reactor vessel construction with an upper plenum. Coolant flows from the upper plenum, down through multiple channels in each of about 600 fuel assemblies, exiting the assembly through a bottom end fitting and into the bulk moderator space. The coolant exits the reactor through six nozzles in the bottom of the vessel and is pumped through light-water-cooled heat exchangers and back into the upper plenum.
- Each of the fuel assemblies is composed of nested, concentric fuel (or target) tubes or annular slugs. The tubes are thin-walled, and the coolant channels between the tubes are thin.

The materials production purpose of these reactors led to unique design criteria and operating characteristics. The basic scientific principles governing the neutron physics, heat transfer, and fluid flow phenomena that occur in Savannah River Plant reactors are those that also apply to all other reactor types. However, the physical conditions and characteristics of the SRP reactors are different from those in power reactors. Thus, Savannah River design, operations, and safety analysis necessitated that unique experimental, theoretical, and computational capabilities be developed, maintained, and applied at the Site. No other sites had similar reactors. Full-scale experimental mockups existed to test the hydraulic characteristics that occurred in fuel assemblies. A full-scale experimental facility representing a one-sixth sector of the reactor was used to model heavy water flow in the moderator space. A full-scale, zero-power experimental reactor (the Process Development Pile, or PDP) was used to measure the nuclear characteristics of full reactor charges. Columbia University performed important thermal-hydraulic experiments for Savannah River, using electrically heated tubes to simulate the nuclear heating that occurs in fuel tubes.[2]

Some History and the State of Nuclear Science and Engineering in 1968

Nuclear science and engineering has a rich personal and technical history including the stories of Marie Curie, Roentgen, Hahn, Strassmann, Einstein, Bohr, Fermi, Bethe, Oppenheimer, and many others. The early history of this science encompassed only a century. With the discovery of nuclear fission just at the advent of World War II, the pace of discovery and development quickened. In his famous letter to President Roosevelt, Albert Einstein advocated pursuit of nuclear research aimed at potential military applications. During the wartime Manhattan Project, some of the world's most talented scientists worked together in theoretical, mathematical, and experimental efforts that led to developing nuclear reactors to manufacture materials that could be separated, purified, and fabricated into nuclear weapons. A number of experimental nuclear reactors were built, and, ultimately, the production reactors were built at the Hanford Works. These reactors produced the fissionable Pu-239 that was used in the "Trinity" test and subsequently used against Nagasaki on August 9, 1945, following by only three days the attack on Hiroshima. Following World War II, nuclear reactor science continued to develop at a rapid pace, with emphasis changing from the startling, dramatic discoveries of the war years to sustained improvement in theories, methods, and nuclear properties of materials. The understanding of nuclear science and engineering that resulted from this work was available and applied in the initial design and development of the SRP reactors. Through the 1950s and 1960s, nuclear reactor scientists and engineers continued to refine this science, and its application, as Savannah River operations evolved and became more effective and more efficient. Important experimental work was performed at Savannah River Plant, but the use of computational and numerical methods was severely limited because "computing machines" included only graphs, hand calculations, slide rules, electromechanical calculators, and large, lethargic vacuum tube-based computers.[3]

Some History and the State of Computer Science in 1968

After the emergence of the abacus in Asia Minor about 5,000 years ago, it was several centuries until the next significant advance emerged in computing devices. In 1642, Blaise Pascal invented a numerical wheel calculator to add sums up to eight figures long. In 1694, Gottfried Wilhem von Leibniz improved on Pascal's machine with a device of wheels and gears that could multiply. In 1812, Charles Babbage built a machine powered by steam and as large as a locomotive, which would have a stored program and could perform calculations and print the results automatically. In 1889, Herman Hollerith, in search of a faster way to compute the U. S. census, invented a computer using punched cards to store data, which were fed into the machine that compiled the results mechanically. In 1931, Vannevar Bush developed a calculator for solving differential equations that had long left scientists and mathematicians baffled. John Atanasoff and Clifford Berry envisioned and by 1940 developed an all-electronic computer that applied Boolean algebra to computer circuitry.

As with nuclear science and engineering, computer science sustained a dramatic acceleration in World War II. The German engineer Konrad Zuse developed a computer to design airplanes and missiles. The British applied computers to breaking secret codes. In 1944, Howard Aiken led a Harvard-IBM team to produce an all-electronic calculator,[4] the Mark I, to create ballistic charts for the U.S. Navy. The ENIAC,[5] also spurred by the war effort, was a vacuum-tube-based computer that was more than 1000 times faster than the Mark I. In 1945, John von Neumann led the development of the EDVAC computer that represented a profoundly important, new computer architecture with a memory to hold both data and a stored program and a central processing unit, which allowed all computer functions to be coordinated through a single source. UNIVAC I, built by Remington Rand in 1951, was the first commercially available computer to take advantage of von Neumann's new architecture. In 1948, the invention of the transistor[6] radically changed computer developments. By the early 1960s, almost all new computers used transistors rather than vacuum tubes and contained printers, tape storage, disk storage, memory, operating system, and stored programs. By 1965, most large businesses routinely used "modern" computers such as the IBM 1401 to process financial information. These machines used new high-level languages such as COBOL and FORTRAN to develop an ever-broadening array of financial and technical applications.

From Site startup in 1951, Savannah River scientists and engineers had only limited access to computers: a Card Punch Calculator 7 was acquired in April 1953; an IBM 650, in 1955; and an IBM 703, in January 1962. In 1966, SRS acquired its first solid-state computer, the IBM 360/65. Punched cards were still used as an input medium, printed paper as the output medium, and, for reasons of cost, magnetic tape was the preferred medium for storing large volumes of data. Random access, hard disk storage was available for rapid access to data in large volumes, but this capability was very expensive. Computer terminals with cathode ray tubes and keyboards were available, but they had essentially no processing capability. They simply transmitted one character at a time from the keyboard to the computer to which the terminal was attached or transmitted one character at a time from the computer to the terminal screen. These computers, as installed, had modest impact on the important work of designing and analyzing efficient and safe nuclear reactors for SRS.

In the early 1960s, other nuclear installations in the United States had acquired "modern" solid-state computers. A number of computer programs had been written to perform some of the basic neutron physics calculations required for nuclear reactor design. In fact, the conceptual "inventors" of the JOSHUA System, H. C. Honeck and J. E. Suich, had both received doctorate degrees in nuclear engineering at Massachusetts Institute of Technology. During their academic work and subsequent work at Brookhaven National Laboratory on Long Island, Honeck and Suich had each authored some of the most advanced computer programs used for calculating neutron physics characteristics that occur in nuclear reactors. Honeck's work, represented by the THERMOS program, described the transport of neutrons as they bounced around within nuclear reactors and interacted with the fuel, moderator, coolant, cladding, and structural materials. One of the most important of these interactions is the "slowing down" (i.e., "thermalization") of "fast" neutrons to lower velocities at which they become more effective in producing a fission event (i.e., the "splitting" of an atom of fuel material) and sustaining the chain reaction. Suich's work represented a significant refinement of this approach. He treated in greater detail and with greater accuracy the "slowing down" of the fast neutrons through the so-called "resonance" velocities, during which the neutrons are particularly susceptible to being captured parasitically by materials in the reactor and thus being removed from contributing to a sustained chain reaction. After Suich's employment at SRS, he and Honeck, during his tenure with Brookhaven National Laboratory and later with the Atomic Energy Commission, collaborated to meld their separate computer programs, plus some new ancillary programs, into an integrated package called HAMMER. HAMMER, although it operated on the "antiquated" computers of the 1950s, became the first significant computational tool for reactor design and analysis at SRS. An important reason for this success was the extensive work at SRS to refine the computational methods and data. Calculated results were normalized to agree with the wealth of measured data from the experimental reactors, engineering test facilities, and from the Savannah River production reactors.

However, even with the availability of HAMMER, the design and analysis of nuclear reactors at SRS was expensive, cumbersome, and very slow. The calculations were performed in a variety of computer programs that were executed in a multi-step, iterative fashion. Each of these calculations was normalized to experimental nuclear, heat transfer, and hydraulic results. Increased understanding of the reactors, coupled with the demand for more detailed reactor safety analyses, led to a highly complex technology requiring a massive system of strongly coupled computational and experimental procedures. New reactor cores were designed, and cores were analyzed during the course of their operation in the SRS reactors. By the late 1960s, the time required from the beginning of a reactor design until its initial operation had increased to almost 18 months. During this period, an average of 150 separate computer jobs had to be performed each month. Considering that there were several concurrent design projects, and that relatively few people were available to lead these design efforts, there was a clear need to simplify the routine use of these programs by better organizing them and better controlling their execution. It became evident in the late 1960s that calculations of this type required a new computing environment, that of the modular data-based system. Thus was the advent of JOSHUA.

Description of the JOSHUA System

JOSHUA is a nuclear reactor design and analysis computational system. Development of the system began in 1968 and continued for more than a decade. The system was developed to perform the extensive nuclear reactor engineering and physics calculations required in the design and analysis of Savannah River reactors with emphasis on operational safety and production efficiency. Subsequent development extended the applications of the system to other, non-reactor areas, including especially the environmental sciences.

JOSHUA Operating System

JOSHUA is a modular, data-based scientific computing system for the multi-step iterative design and analysis of nuclear reactors. JOSHUA consists of an operating system and an applications system. The operating system facilitates data management, program execution, and computer terminal use. The applications system is a collection of computational modules that perform the numerical operations representing the science and engineering models.

What does "multi-step, iterative" mean? Nuclear reactor design requires many computational steps. One such step might be the calculation of the heat produced in one fuel assembly. Another step might be the calculation of the fluid temperature in a coolant channel, as heat is transferred from the fuel to the coolant. Steps are often repeated using different sets of input data, or different models to perform the particular calculation.

Furthermore, groups of steps are often repeated until some reactor design criterion is met. This process is composed of multiple steps (i.e., multi-step), and it involves repeated executions of one or more particular steps (i.e., iterative.)

What do "modular" and "data-based" mean? In the early 1960s when a calculation was performed by a computer program, the input data were punched on cards, and the computed results were printed on paper. If some output data from one step were required as input data to a subsequent calculation, these data were manually transcribed from the printed report to an input card. In the mid 1960s, random access, disk storage devices became available at reasonable prices and thus made it practical to save results from one step to be made available to subsequent steps. When selected output data from a step are placed in a "pool" of data residing on disk storage and made available to all other steps, the resulting system is called a "data-based" system and the computational steps, or computer programs, are called modules. Modules can execute other modules to facilitate development of complex computational procedures, such as those required for nuclear reactor design and analysis.

From the beginning, it was planned that JOSHUA would make use of named data records. One collection of material property data required by reactor calculations is the set of neutron fission cross sections for each of multiple neutron energy groups in the U-235 isotope. Prior to JOSHUA, such data might be stored on magnetic tape that was read by the computer as tape unit 20, record 12,456. Although perfectly satisfactory to the computer, this name had no meaning to the reactor designer. With JOSHUA, these data were stored on a random-access disk in a record named MULTIGRP.U235.FISSION. JOSHUA allowed records of up to 16 names of no more than 8 alphanumeric characters in length, separated by periods.[8] The resulting database can be represented as a hierarchical, tree structure, and JOSHUA implemented a relatively sophisticated data management system to facilitate data access, data modification, data creation, access security, search facilities, and other important data management capabilities. These data management capabilities served the computer programs, which were written in an extension to the FORTRAN programming language, and human users of computer terminals.

Also from the beginning, it was planned that JOSHUA would make extensive use of computer terminals as the principal input/output interface between the computational system and the reactor design engineer. Use of computer terminals provided for the following:

- Entry of data directly into the database
- Inspection of results in the database
- Modification of data in the database
- Execution of modules (i.e., computer programs comprising the computational steps)

These capabilities are important in the reactor design process. They allow the reactor designer to dynamically control the database and the computational sequence that is performed, without resorting to using punched cards and printed reports, or other antiquated media. This greatly reduces the time required for a reactor designer, or a group of designers on a team, to go through a complicated computational process.

JOSHUA Applications System

The task of nuclear reactor design and analysis is basically one of simulating mathematically the state of the nuclear reactor in static and transient situations. Reactor design considers the static case, and reactor safety analysis considers the transient case. The “state” of the nuclear reactor is defined as the space- and time-dependent parameters primarily associated with power density (e.g., heat generation rate), neutron density, temperature of solid and fluid materials, density of solid and fluid materials, and fluid velocity. All of these state parameters have different values at each location within the reactor (i.e., the “space dependence”) and at each point in time. Further, the neutron density depends also on the neutron velocity, since the density of “slow” neutrons at a point in space and time may be different than the density of “fast” neutrons at the same point in space and time. Neutron density, temperature, material density, and fluid velocity are described mathematically by a set of differential equations. These equations are coupled because the coefficients in the equations, representing collections of material properties (e.g., neutron cross sections, heat capacities, thermal expansion coefficients), are themselves functions of the state parameters, especially temperatures and densities. The modules that comprise the JOSHUA applications systems are a collection of computer programs, which employ the JOSHUA system facilities used by the reactor designer to solve the aforementioned sets of equations. The overall design problem is broken down into smaller component problems that balance solution accuracy with solution cost and permit the designer to approach the overall design problem in smaller steps.

The heart of the applications system is comprised of modules that model the nuclear characteristics of the system and of other modules that model the engineering characteristics of the system. The nuclear modules are of two basic types. One set of nuclear modules uses the methods of integral transport theory (or alternative transport theories, such as Monte Carlo or response functions) to compute the neutron densities in one, or a few, reactor assemblies. These transport calculations are performed as static calculations in two-dimensional space representing a horizontal plane within the assembly. The other set of nuclear modules uses the methods of neutron diffusion theory to compute the neutron densities throughout the entire reactor. These diffusion calculations are performed as either static or dynamic calculations in one-, two- or three-dimensional space. Both sets of nuclear modules perform the calculations under an assumed or given set of engineering parameters (e.g., temperatures and densities). The engineering modules use an assumed or given distribution of power densities (i.e., heat generation rate) and compute engineering state parameters (e.g., temperatures and densities) in the metal and coolant of a reactor assembly and throughout the moderator space of the reactor. These calculations can be either static or dynamic, and they are inherently three dimensional.

In addition to these nuclear and engineering modules, there are ancillary modules that are used to process nuclear, thermal, and hydraulic properties required in the calculations. This processing treats the dependence of neutron cross sections on temperature and density, for example. Other modules are used to process input data (e.g., reactor geometry, fuel concentrations, moderator purity) to select amongst various calculation options and to prepare output reports of calculated results.

These basic and ancillary modules can be combined in iterative, multi-step calculations for a variety of design and analysis purposes. The JOSHUA operating system facilities permit the development of special control modules, which direct the execution of the basic nuclear, engineering, and ancillary modules, for the wide variety of design and analysis purposes required. Some of the more important of these design and analysis capabilities are listed below:

- Assembly design. This calculation supports the selection of fuel and target tube dimensions and material compositions for a particular reactor assembly design.
- Charge design analysis. This calculation is used to predict the normal operating characteristics as a function of fuel depletion throughout the life of the reactor core.
- Flow-zoning analysis. This calculation is used to distribute coolant flow among assemblies within a particular reactor design.
- Confinement protection analysis. This particular safety analysis predicts the pressure surge in the reactor confinement system for a variety of hypothetical accidents in which the safety system fails. The results are used to design charges and define operating limits that ensure that the reactor confinement system will not be breached.
- Thermal-hydraulic limits analysis. This particular safety analysis predicts the transients in assembly effluent temperature that would result from hypothetical accidents terminated by automated safety systems. Analyses of these accidents are used to define limits on normal operating temperatures.
- Film-boiling burnout analysis. This particular safety analysis predicts the extent of fuel damage due to film-boiling burnout under static and transient conditions. These analyses are used to define limits on the heat flux at the fuel-coolant interface. Quantitative analysis of the phenomenon is based on experiments from which the degree of damage can be corrected with Burnout Safety Factor, the ratio of burnout heat flux to the actual heat flux.

The Results, Significance, and Impact

The JOSHUA System was on the leading edge of modern, scientific computing technology. As one millenium ends and another begins, it is difficult to recall the state of technical computing in 1968 when the development of JOSHUA began. In the subsequent three decades, we have seen a revolution in computing. During the last two decades of the twentieth century, the cost of computing power decreased about 30% annually and microchip performance power doubled every 18 months. Computing power costs have fallen more than 10 millionfold since 1968. The JOSHUA system could not have been built a decade earlier because computer hardware simply did not exist to support the data management, computer terminals, and other facilities provided by the system. Further, the computational power required for the nuclear reactor design and analysis calculations was prohibitively expensive even for national defense purposes. Had the JOSHUA system been built three decades later, it would have made full use of modern servers, workstations, personal computers, data management systems, and the Internet. The system development would have used "off the shelf" technology and would have required considerably less ingenuity and innovation. Its development would have required much less time, at much less cost than was the case in 1968. One impact of JOSHUA is that it demonstrated to computer scientists the utility and power of the computational facilities that the system supported.

JOSHUA incorporated into a single, modular system the very best of nuclear physics and engineering models available at the time. The utility and power of such a comprehensive design and analysis tool was demonstrated to the entire nuclear industry and influenced the development of nuclear physics and engineering models at other sites.

Another, and perhaps the greatest, significance of the JOSHUA system is that its application significantly enhanced the efficiency, effectiveness, accuracy, and safety of nuclear reactor design and analysis capability at Savannah River Site.

Contributors

Many individuals contributed directly and significantly to the design, development, and application of the JOSHUA System. Their success was built upon the brilliant work of technology pioneers in nuclear, thermal, and hydraulic science and engineering. This paper has attempted to reflect the appropriate credit to early pioneers and to the early Savannah River Plant people. All of them were an important part of the legacy that led to JOSHUA.

Singling out an individual contributor risks overlooking important work of others. However, in the case of the JOSHUA System, three individuals should be acknowledged for their exceptional contributions. Dr. John E. Suich and Dr. Henry C. Honeck were responsible for the initial concepts of the JOSHUA System, both in the operating system and in the applications system. Their engineering and computational insight, vision, and commitment inspired the effort. Suich and Honeck were most fortunate to find themselves in an organization that at the time was led by J. W. Croach, Technical Director of DuPont's Atomic Energy Division. Mr. Croach possessed a solid understanding of physics, engineering, and computer science. He understood the need for a modular, data-based system such as JOSHUA, and he provided the executive support and encouragement.

To those of us who had the pleasure of working in the presence of such talent, we remember these three leaders with continued appreciation and admiration.

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Endnotes

1. The name JOSHUA is not an acronym and was chosen largely arbitrarily by H. C. Honeck and J. E. Suich soon after they conceived the system. Suich had developed an earlier program that he named JERICHO. The name JERICHO led to the name JOSHUA from the Biblical story of Joshua at the battle of Jericho, where the blowing of trumpets and the shouts of the people caused the city walls to crumble. When a JOSHUA computer program failed, an error message was printed, "And the walls came tumbling down." After JOSHUA development had progressed for several years, Honeck and Suich sponsored a contest among the system developers for the cleverest acronym associated with the name JOSHUA. Propriety prevents the reporting in this paper of the winning suggestion.
2. The thermal-hydraulic experiments at Columbia University used large amounts of DC electrical power to simulate nuclear heating in mock fuel assemblies. The story is told, perhaps apocryphally, that these experiments were conducted late at night because the New York subway system also used large amounts of DC power and conducting the experiments during daylight hours would have overloaded the electrical system!
3. We sometimes felt that the computers we were using contained as much steel as the reactors we were trying to analyze, and that

the computers generated almost as much heat as the reactors.

4. The Mark I, built in 1944 by a joint Harvard-IBM team, was an electronic relay computer which used electromagnetic signals to move mechanical parts. The computer was about half as long as a football field and contained about 200 miles of wiring. It required 3-5 seconds per calculation in a sequence of calculations that could not be changed.
5. The ENIAC consisted of 18,000 vacuum tubes, 70,000 resistors, and 5 million soldered joints. It consumed 160 kilowatts of electric power but had computing speeds more than 1,000 times faster than the Mark I.
6. Although transistors were clearly an improvement over vacuum tubes, they still generated significant amounts of heat. This problem was solved by the development of the integrated circuit in 1958 by Jack Kilby, an engineer at Texas Instruments. This development was continued with more and more electronic components being packed onto a single chip. In the 1970s, large-scale integration (LSI) permitted hundreds of components on one chip. In the 1980s, very large-scale integration (VLSI) extended this to hundreds of thousands of components on one chip. And by the late 1980s, ultra large-scale integration (ULSI) extended that number to the millions.
7. The Card Punch Calculator or CPC was an electro-mechanical device in which the user inserted one punched card in order to simulate the actions of an adding machine!
8. Rather than "periods," we should have called them "dots" as in "JOSHUA-dot-com."
9. There is only one reference given in this paper. That reference contains a complete description of the JOSHUA System as of 1975, plus a complete list of published reference materials from 1968 through 1975 regarding the development of the JOSHUA System.

Acknowledgment

The author gratefully acknowledges the many comments, suggestions and careful reviews of this paper by his colleagues M. R. Buckner, G. F. Merz, T. F. Parkinson, and D. A. Ward.

Biography

During his undergraduate studies, John Stewart was employed as an engineering co-op student with Du Pont at the Savannah River Plant and Laboratory in Aiken, South Carolina. During his graduate studies, he was a part-time employee of Oak Ridge National Laboratory. He was granted B.S., M.S., and Ph.D. degrees in nuclear engineering by the University of Tennessee. Following graduate studies, John was employed in 1969 as an engineer at the Savannah River Laboratory and assigned to reactor design, analysis, and computational modeling work. There, he participated in the development of JOSHUA, a comprehensive computational system for design and analysis of nuclear reactors, in both technical and supervisory roles. In 1975, Mr. Stewart was granted a leave-of-absence to accept a one-year appointment as a visiting research associate at MIT, leading the development of a major power reactor safety analysis computer program. In 1976, he returned to Savannah River and held a variety of management positions in research, development, engineering, manufacturing, and employee relations in the Laboratory and Plant. In 1984, he was named Technical Manager of the Atomic Energy Division in Wilmington, Delaware, and in 1987, he was named Design Manager in DuPont Engineering-Atomic Energy Division. In 1988, he became a product development manager in the Electronic Imaging Division. John returned in 1990 to Du Pont Engineering and served as manager of energy engineering and leader of the corporate energy conservation program until his retirement from Du Pont in 1998. He currently lives in the Atlanta area.

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Discovery That Nuclear Fission Produces Tritium

Edward L. Albenesius, J. Henry Horton
Harold M. Kelley, Daniel S. St. John,
and Robert S. Ondrejcin

Abstract

Webster defines serendipitous as 'the finding of valuable or agreeable things not sought for'. This definition truly fits the surprising scientific discovery made early in the operation of the Savannah River Plant that tritium is a product of nuclear fission. Entirely by chance in 1957 in a hydrologic tracer study with tritium at a waste water disposal site, it was discovered that tritium was present in very measurable amounts in irradiated nuclear fuel. The tritium-in-fuel phenomenon remained unexplained for two years until (again by chance) it was proposed to apply its presence as an analytical tool to measure reactor exposure of nuclear fuel. This idea, which quickly turned out to be impractical, caught the attention of the research staff at what was then the Savannah River Laboratory and a basic scientific inquiry was eagerly encouraged. An all-out study was completed in the remarkable span of ten weeks, ending in the successful discovery of an entirely overlooked portion of basic nuclear data.

Introduction

With the advent of the huge research programs needed to develop the atomic bomb, chemists and physicists had a great time discovering and characterizing the hundreds of byproducts created in the process of nuclear fission. By the late 1940s, it was assumed that all radioactive fission products with half-lives greater than a few seconds had been identified. In these studies, attention focused logically on the products of binary fission: ternary fission was recognized as a curious source of helium as energetic alpha particles. A few studies sought other light particles without notable success. No speculation existed that tritium might be formed this way; however, if anyone had looked, not by examining individual fission events as a physicist would, but by looking at the accumulation of trillions of events, as a chemist would, it would have been an easy discovery. Tritium has a moderate half-life of 12 years, is easy to separate chemically from interferences, and can be measured precisely. By the late 1940s, research attention had shifted away from 'old hat' fission phenomena to the formation of new elements by neutron and charged particle addition.

Discovery of Tritium in Fuel Reprocessing Waste

At the beginning of operation of the Savannah River Plant's (SRP) two irradiated-fuel reprocessing plants, lightly contaminated condensate from waste evaporators was discharged into earthen seepage basins. The Health Physics Section (HP) was developing an understanding of the movement of these radioactive contaminants into the groundwater by monitoring a series of nearby wells. Monitoring data from the wells was pretty barren for ionic fission products because the soil, as expected, was sorbing these products. Henry Horton, a specialist in soil science, and myself (Ed Albenesius), an organic chemist, ran this program. We had a crude model of groundwater movement based on hydrology, but we were eager to confirm data in the field. So we came up with the bright idea of 'spiking' a well with a small amount of tritiated water that would move freely with the groundwater. We would then measure its arrival behavior at nearby downslope wells. We spiked a well in December 1956, and, to our surprise, a week later the first sample of the nearest downslope well, 20 feet away, contained tritium, a rate of movement 10 times what we expected. To our complete astonishment, a hurried sample of a well 100 feet away also contained tritium! We then rushed back to the seepage basin and discovered that it was the tritium source; subsequent careful analysis of all the waste streams feeding the basins traced the source to the process vessel in which the uranium fuel was dissolved to start the separations process. The presence of tritium was also confirmed in the enriched uranium fuel waste.

Over the next two years, as we strove to absorb the significance of this discovery, the reality was that we now had an extraordinary tool to understand the hydrology of the seepage basin system. With a perfect tracer leading the way, numerous soil cores were taken to optimize the placement of monitoring wells and to develop seepage basin hydrology in three dimensions; these steps enabled credible projection of future basin performance. In all, over 200 monitoring wells were installed over the next 10 years.

Lost in the shuffle for the first two years was any effort to understand scientifically why the tritium was present in the irradiated fuel. Off-the-cuff speculation tended to dismiss its presence as possibly due to a lithium impurity in the fuel (lithium splits into tritium and helium in a nuclear reactor). Or the tritium diffused into the fuel from the reactor heavy water moderator, where it is an impurity that grows in by neutron capture in deuterium. Or perhaps it might be a contaminant related somehow to the large amounts of reactor irradiated lithium-aluminum target rods used in the Site's large-scale tritium production process. Surely, the casual thinking went, with all that tritium on the Site, it should not be too surprising to see some turn up in unexpected places such as waste basins. The turn to scientific investigation of the actual source of the tritium and the completion of this whole serendipitous process required one more unplanned event.

How the Scientific Study Began

Two years after the discovery of tritium in the fuel separations waste, I was transferred from the Health Physics Section to the Analytical Chemistry Division of the Technical Division, the research arm of the Site. The environmental chemistry emphasis of the old assignment instantly supported the process chemical research in the new assignment. An intriguing problem at the time was the measurement of burn up (consumption of nuclear fuel in various reactor configurations). Among existing methods of burn-up determination, isolating and analyzing a specific fission product, such as Cs-137, was most frequently employed. These methods were difficult analytically and yielded imprecise data.

At the time of the discovery of tritium in the waste from the fuel separations plants, calculations indicated a rough correlation of tritium quantity with reactor exposure of the fuel (about 1 tritium atom per 40,000 fissions). The idea now surfaced that if correlation could be proved and precisely measured, the tritium in nuclear fuel could offer a useful approach to determining burn up because, analytically, tritium (as tritiated water) is easy to isolate and precisely measure. I took my idea to Harold Kelley, my research manager, and proposed a feasibility study. Kelley's reaction was one of unbridled enthusiasm. Hearing of the tritium-fuel finding for the first time, Kelley sensed that a potential scientific discovery was on the doorstep. At Kelley's urging, I discussed the tritium data with one of the staff's reactor physicists (Dan St. John) seeking a clue that might link tritium with fission phenomena. St. John knew of no such link for tritium, but pointed out that the helium ion is known to occur in a rarer mode of fission known as ternary fission. This was an extremely valuable lead that provided a focus for thorough search of the scientific literature on fission phenomena.

Search of the Ternary Fission Literature

No report could be found in the literature citing evidence for or against the formation of tritium in the process of nuclear fission. The phenomenon of alpha particle emission in ternary fission had been extensively studied. The mechanism had been well established. No evidence was found for the formation of tritium or deuterium. However, in one study of the energy distribution of fission alpha particles, the appearance of tritons was noted but was dismissed as an inconsequential interference. Studies of proton (the third hydrogen isotope) formation in fission were carried out by several authors. Protons were observed in a study with photographic emulsions but were discounted as collision products. Protons were also observed at a frequency of 1 in 5000 fissions in another study with a carbon dioxide range chamber. However, a theoretical study stated that proton emission should be practically forbidden in comparison to alpha emission. Thus, the literature had no references on the formation of tritium in fission and had conflicting evidence for the proton. The conversation with St. John and the resulting focus of the literature review had now taken the study from concept to hypothesis. What remained to be done was to prove the hypothesis in the laboratory.

Proof that Nuclear Fission Produces Tritium

The laboratory experiments to eliminate speculation as to alternative non-fission sources of the tritium and, if successful, to measure precisely its yield in the fission process required the skills of an accomplished radiochemist. Such a person was Bob Ondrejcin, who stepped forward to design and conduct these exacting studies (Albenesius 1960).

Three experiments were carried out to provide proof that the radioactive species being measured was tritium, that the presence of tritium was not due to irradiation of a lithium impurity in the uranium fuel, and that the tritium did not come from the heavy water moderator of the reactor. The experiments were as follows:

- A sample of water from the acid solution in which irradiated uranium had been dissolved was converted to hydrogen, and the gas was diffused through a palladium barrier. The diffusate was reoxidized to water, and the expected tritium content was verified.
- The lithium content of the uranium fuel was determined. A sample of uranium from a typical fuel element was dissolved in nitric acid, and the uranium was precipitated. The supernate, which contained any possible lithium impurity in solution, was analyzed by emission spectrography. The lithium content was less than 6% of the amount required to produce the level of tritium observed in irradiated uranium.
- A sample of enriched uranium was irradiated in a graphite-moderated experimental reactor that contained no heavy water. The sample was dissolved in nitric acid, and the water was separated by distillation. The ratio of tritium to fissions was comparable to the ratio in fuel irradiated in heavy-water-moderated systems.

For the precise measurement of the yield of tritium in fission, eight sections were cut from rods of uranium irradiated in an experimental fuel assembly in an SRP production reactor. Total fissions in the assembly were calculated from measurement of coolant flow, temperature, and neutron flux. The cut sections were cleaned of any adherent film of tritiated deuterium oxide, then dissolved in nitric acid in an apparatus designed to collect the offgas. About 25% of the total tritium was found in the gas phase. The overall yield of tritium was calculated to be 1.05 +/- 0.09 atoms per 10,000 fissions.

Following publication of the discovery of tritium as a product of fission (Albenesius 1959), two independent studies in 1961 confirmed tritium emission in the spontaneous fission of californium-252 and measured its energy distribution. These works also proved our hypothesis that the tritium was formed as the triton in ternary fission. Emission of the triton in the fission of uranium was also independently verified in 1963. The fission yield of tritium for uranium was also independently confirmed in 1962.

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Biographies

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Degrees:
College of Charleston 1944-47
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1951-53 Research Chemist, Health Physics SRP
1953-59 Supervisor, Environmental Monitoring and related programs
1959-64 Research Supervisor, Analytical Chemistry Division, SRL
1964-87 Research Manager, Managed a series of SRL programs in support of process development, environmental chemistry, and low-level and transuranic waste management.
1988-89 Assigned to DOE HQ; Coordinated revision of DOE Order 5820.2A Radioactive Waste Management
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1970-85 Laboratory Director, Explosives Department, at the Experimental Station. During this period, the Explosives Department Laboratories were combined with Nylon research to form the laboratory for the new Petrochemicals Department. Research areas included simulations and reaction kinetics for the production of acrylonitrile and statistical analysis of stratospheric ozone measurements
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1951-52 Jr. Chemist, Argonne National Laboratory
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Savannah River Site Canyons—Nimble Behemoths of the Atomic Age

LeVerne P. Fernandez

Abstract

Processing extremely radioactive materials for nuclear weapons is accomplished at Savannah River in chemical separations facilities called “canyons”. These large, remotely operated, heavily shielded facilities were constructed early in the Cold War to provide raw material, initially plutonium-239, for the U.S. nuclear arsenal. As the chemical separations processes developed and matured, these canyons showed a versatility not in their original scope. They were used to process a variety of materials in ways not envisioned by the original designers. With some renovation or revision, these facilities have been used to support the needs of the United States for other radioactive materials. Savannah River canyon facilities provided the raw material, neptunium-237, which was irradiated at the Savannah River Plant (SRP) to produce plutonium-238 used to power U.S. space probes that go where solar panels are too weak to suffice. Though almost 50 years old, the canyon facilities still function well, a real tribute to the original designers, builders, and the subsequent remodelers. Significantly, they have also operated without a nuclear criticality incident.

Introduction

The chemical separations plants of Savannah River have had an interesting and proud history. This paper about the flexibility of these huge concrete and steel chemical processing plants is only a part of that story.

Original Scope of Facilities

Initially, the Site’s two separations plants were designated Building 221-F and Building 221-H and are commonly called, respectively, F Canyon and H Canyon. They were designed to separate weapons-grade plutonium (Pu-239) from irradiated uranium. In the beginning, the heavy-water-moderated reactors at SRP used natural uranium for fuel and target elements. F Canyon and H Canyon were designed to be identical to provide redundancy in case one was lost for any reason.

In 1950, when the U.S. government asked Du Pont to construct and operate the Site, Du Pont had an extensive engineering staff and vast construction capabilities. Also, Du Pont had already constructed and operated the Hanford Works in Washington State, which produced much of the weapons material in the early days of the nuclear age. At Hanford in 1944-45, Pu-239 was separated and purified by a precipitation process. By the start of construction of the Savannah River Plant in 1950, work at Oak Ridge National Laboratory, Knolls Atomic Power Laboratory (KAPL), and Argonne National Laboratory, among others, showed that separation could be achieved successfully in two-phased liquid systems. A countercurrent flow of an acidic aqueous stream with an immiscible organic phase containing a complexing extractant was selected for Savannah River. The process was called Purex and used nitric acid in the aqueous phase and tri-n-butylphosphate (TBP) dissolved in a lightweight organic liquid, such as n-paraffin, for the organic phase. SRP selected mixer-settlers for this operation, while Oak Ridge selected pulsed columns. Both mechanical configurations were successful and have been the subject of many reports (Joyce 1959; McKibben et al. 1979; McKibben 1989; Orth 1964a; Orth 1964b; Orth and McKibben 1969; Orth and Olcott 1963).

The SRP canyons were designed to be operated and maintained remotely to minimize worker exposure to radiation. These structures are called canyons for the long, narrow spaces in which processing takes place. The processing portions are segregated into a hot canyon with very heavily shielded protection and a warm canyon which, while still radioactive, requires less shielding. The canyons are parallel to each other and are separated by the shielded and inhabited parts of the buildings. All the radiochemical operations, including the receipt of targets, dissolving, feed clarification, solvent extraction (and later, ion exchange), and waste handling are performed remotely in these concrete-shielded buildings.

Each building is 255 m (850 ft) long, 37 m (122 ft) wide, and 20 m (66 ft) high (Starks 1977). Each canyon consists of 17 13.1-m (43 ft) sections and a 25.9-m (85 ft) section. Each canyon has 12 processing sections with each section having four cells. Each cell is capable of receiving a piece of equipment and siting it, via trunions, in a specific known location. The pattern of embedded piping is duplicated over each of the 12 identical sections; that is, each section is an exact replica of all other sections in locations of tank positioning guides and wall nozzles. The precision of this replication is such that equipment that fits in one location will fit in another similar location to within 1/16-inch. Each section of the canyon with its imbedded piping was constructed as one continuous concrete pour.

Much of the flexibility of these canyons can be attributed to the remarkable quality of the work of the craftsmen who built these behemoths. All the services (electrical, steam, hydraulic fluid, and chemical transfers) are available from piping at every processing section. The high-quality construction along with an accessible "cold" (non-radioactive) mockup shop section guarantees that new or replacement equipment will fit. This has contributed greatly to the canyon flexibility and undoubtedly reduced the amount of solid waste generated by canyon operations. In fact, being able to design and install different equipment that utilizes the previously installed services has enabled increased performance under the original flowsheet from 3 metric tons uranium per day to 15 metric tons per day. It has also allowed the installation and use of equipment not visualized in the original design, as this paper will illustrate.

Equipment is moved or replaced in the canyons by means of overhead cranes. In the hot canyon, the operator is shielded from direct radiation by a thick concrete wall and must view the operation through an optical arrangement. Originally, these optics were of early submarine periscope style. In the mid 1980s, the canyon cranes were replaced, and the optics were upgraded to include a bevy of remote television cameras, so the view can be not only shared but also recorded for instructional or any other purposes.

Waste Handling

The solid waste from the canyons themselves is almost completely either damaged or obsolete equipment. This equipment, whether jumpers, tanks, evaporators, mixer-settlers, or ion exchange columns, was decontaminated, wrapped, boxed, and transported to the Site's burial ground.

The liquid waste from canyon operations had several components (Starks 1977). The non-contact cooling water used to control the temperature in some tanks and provide cooling water to evaporator condensers and other non-contact cooling operations exited the canyons to a diversion canal with monitoring, which was initially periodic, then continuous as monitoring capabilities improved. This stream also contained the condensate from steam used in heating coils in process operations in the canyons. Non-contaminated water was discharged to plant streams, which eventually flowed to the Savannah River. In the event contamination was detected, the water was diverted to a lined holding basin for cleanup prior to discharge. Though not a frequent event, the holding system was used on several occasions, usually as the result of a failed steam coil in an evaporator. The system successfully prevented significant radioactive releases to the environment.

The radioactive liquid waste from the canyons was released from the canyon as high activity waste. It contained the long-lived fission products and the dissolved aluminum cladding from both fuel rods and targets. This material was transferred to large underground steel storage tanks for later treatment. This body of waste eventually reached some 40 million gallons and is currently being converted through a vitrification process to a solid waste form for eventual geologic disposal. The low activity liquid waste was evaporated to recover nitric acid wherever practical and to reduce volume. The reduced waste is now being converted to a solid form, saltstone, and stored onsite in bunkers.

Ventilation and Contamination Control

The air system in the canyons operates on directed air flow and pressure differentials. Building space that is occupied or could be occupied by people is fed chilled air from an external source. This air is heated as needed and maintained at slightly lower pressure than the outside air pressure. The process area operates at air pressure below that of the personnel space so that contaminated air does not escape into the personnel space.

Some processing facilities within the canyon buildings are not in the canyons themselves. These are areas where the desired radioactive product, such as U-233, Pu-238, or Pu-239, is converted to solid form. Because these radioactive materials pose serious alpha particle contamination threats but much smaller beta-gamma radiation threats, these processes are usually carried out in gloveboxes. In these process areas, air flows from the "uncontrolled" personnel area to the process area to the glovebox, so that the flow of air is always toward the radioactivity and works to prevent the spread of contamination. In the canyon process area of the building, which operates at air pressure negative to the office and working spaces, all process tanks are connected to a vessel vent system that operates at a pressure below that of the general canyon process space. All the canyon gases are filtered through a large and elaborate sand filter before discharge through a tall exhaust stack. This system has been extremely effective in controlling the release of radioactive solids to the environment. Radioactive gas release was minimized by extending the cooling time of the fuel to be processed to allow time for radioactive decay of xenon and I-131 and by using silver absorbers to remove iodine.

Purex Solvent Extraction

As mentioned earlier, both canyons were designed and initially started up using the Purex process. This process has seen many variations. The TBP in the solvent has varied in different applications from 3.5% (ref) to 50% (ref) with 30% being most common (Orth 1965). Purex has been used to process fuels and targets varying in U-235 from depleted (~0.2%) to over 93%. It has also been used to process highly irradiated plutonium, recovering plutonium as well as actinides with higher atomic numbers.

A modification of the Purex process called Thorex, developed at KAPL and the Savannah River Laboratory (SRL), has been used to recover U-233 and thorium from irradiated thorium targets.

Np-Pu Ion Exchange Frames

As the enriched portion of the reactor fuels increased from natural to about 93%, the enriched uranium that was recycled began to increase in U-236. Further irradiation of this U-236 produced neptunium, specifically Np-237, which in turn was recovered and further irradiated to produce Pu-238. This isotope has been used as a power-generating heat source to provide onboard electric power for satellites sent into deep space.

When the need arose to process highly radioactive materials where the quantities were less than a ton, the solvent extraction system designed for metric tons per day was simply too large for the task. The holdup in the solvent in the mixer-settlers alone is about 5000 gallons. Sometimes, ion exchange processes have advantages over solvent extraction, especially so in batch operations where the quantities of material to be handled are small, relatively, and the desired product concentrations are high.

After splitting out the Np-237 in a modified Purex flowsheet, it was recovered and purified on ion exchange columns. To accomplish this, a small-scale, by industrial standards, ion exchange processing plant was constructed on three 10 x 10 x 17-ft steel frames, which were set in a canyon cell and connected to appropriate services (Mottel and Proctor circa 1963). The equipment system included a dissolver, 8 ion exchange columns, and 16 solution adjustment and collection tanks complete with instrumentation and solution transfer devices. Each frame, with its complement of tanks, columns, and piping, was capable of being installed or removed as a single unit. Only those few items subject to periodic failure could be removed individually from the frame, while the majority of the equipment, including the tankage and piping, were permanently fastened to the frame structure. These frames were fed, serviced, controlled, and sampled remotely from common canyon services and connections, albeit, some interesting modifications were employed; for example, six 1/2-inch tubes were pulled as a bundle through a single 3-inch pipe to permit flow control and sampling of the ion exchange columns.

These frames, certainly not envisioned by the original designers, processed irradiated neptunium targets to provide the desired Pu-238 and recover the residual Np-237 (Poe et al. 1963). Plutonium-238 from this effort provides heat, which via thermoelectric conversion to electric power, has provided on-board power for U.S. space exploration vehicles including the original Viking Explorer, the Voyager series, and, more recently, the Magellan spacecraft to Jupiter.

Electrolytic Dissolver

Just as in the case of the frames, the flexibility of the canyons was demonstrated once again in the need at SRP to process non-SRP fuel. These fuels from various federal programs were clad with either stainless steel or various zirconium-based alloys. These claddings are not amenable to dissolution in a nitric-acid-based system. Also, the safety of the existing process vessels and piping, which are constructed of stainless steel, would be severely threatened by any chemical process that would dissolve stainless steel cladding or zirconium cladding. After design and testing, an electrolytic dissolver utilizing a liquid cathode concept was installed in H Canyon. Here again, the preciseness of a canyon location coupled with normal canyon services augmented by a special DC power connection capable of 25 volts and 10,000 amps enabled installation and use of equipment previously demonstrated in cold pilot plant studies. This dissolver operated successfully through five campaigns over a span of ten years. After dissolution, the fuels were successfully processed by a normal Purex flowsheet.

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Acknowledgments

I have been fortunate to be involved with several very talented teams in my assignments. Also, I had as co-workers and supervisors some of the very finest technical minds, which make my memories of the years at Savannah River very satisfying. I am grateful for the experience.

Biography

LeVerne P. Fernandez obtained his Ph.D. from the University of Virginia. He worked over 35 years at Savannah River with the majority of the time in technical support of the chemical separations processes, especially HM and Purex flowsheets, the electrolytic dissolver, and frame operation for Np-237 and Pu-238 recovery. The technical support for the first General Purpose Heat Source Pu-238 heat source capsules fell to his group. He led the group that designed and built the M-Area Liquid Effluent Treatment Facility and the M-Area Air Stripper for the removal of volatile chlorocarbons. These facilities made possible the closing of the M-Area Seepage Basin and the restoration of Lost Lake.

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Development and Performance of Centrifugal Mixer-Settlers in the Reprocessing of Nuclear Fuel

Albert A. Kishbaugh

Abstract

An 18-stage centrifugal mixer-settler, designed and developed at the Savannah River Laboratory (SRL), successfully separated plutonium and uranium from radioactive fission products at the Savannah River Site (SRS). This paper discusses the design of the mixer-settler and tests that were conducted before this equipment was installed at SRS and subsequent performance with radioactive solutions. Advantages of the centrifugal equipment over the pump-mix mixer-settler are associated principally with the fiftyfold reduction in liquid volume holdup, and include reduced exposure of the solvent to radiolytic degradation, increased nuclear safety, easier flushing, and greater operating flexibility. The unit has a capacity of over 60 gpm total flow.

In the new mixer-settler, pumping, mixing, separating, and decanting are all performed in a single device with a single rotating shaft. A particular feature of the new mixer-settler is the use of air pressure in a weir chamber to regulate remotely the emulsion position in the centrifuge. The centrifugal stages have high mass transfer efficiency; for example, an overall efficiency of approximately 95% is attained in stripping uranium from solvents. The unit has performed well since start up in October 1966. Maintenance requirements have been small, decontamination and losses are satisfactory, the solvent picks up less then one-fifth as much gamma activity as it did in the pump-mix mixer-settler, and shutdowns and startups are made rapidly without extensive flushing.

Introduction

Uranium and plutonium are recovered from irradiated actinide fuels and targets predominantly by extraction processes using kerosene-diluted tributyl phosphate as a solvent. Radiation from fission products damages the solvent and decreases the separation efficiency of the process. Because the radiation damage to the organic solvent varies with the time that the solvent is exposed to ionizing radiation and because this exposure time is determined primarily by the time required to separate the aqueous and organic phases, the use of centrifugal settling should clearly be superior to gravity settling.

The centrifugal extraction equipment available commercially was not designed for use with radioactive solutions and was not amenable to the remote maintenance procedures required for such service. Consequently, the Savannah River Laboratory (SRL) developed a centrifugal mixer-settler that met these requirements (see references). Three 6-stage assemblies were built, tested at a non-radioactive semiworks facility, and installed in SRS as part of the Purex process. The three assemblies were joined into an 18-stage bank that replaced a 24-stage bank of pump-mix mixer-settlers used for co-decontamination of uranium and plutonium from fission products. These centrifugal mixer-settlers were expected to have the following advantages relative to the pump-mix mixer-settler:

- Reduced exposure of solvent to radiation
- Reduced aqueous and solvent inventories
- Reduced space requirements
- Easier flushing for process changes
- Greater safety in handling fissionable materials
- Accommodation to a wide variety of process solutions with varying densities and viscosities

Design and Mode of Operation

Each stage of the centrifugal mixer-settler, shown in Figure 1, has a 5 HP, 1745 rpm motor with a vertical, overhung shaft to which is attached a 10-inch-diameter separating bowl and, at the bottom, a mixing paddle-pump. Both the heavy (aqueous) and light (organic) phases flow by gravity from adjacent stages and enter the pumping-mixing chamber through the tee at the bottom. The phases are mixed by the paddle and ejected at the periphery of the mixing chamber into an upper chamber where the mixture moves inward along antivortex vanes. The vanes convert some of the rotational energy to pressure so that the mixture can be injected into the bottom of the rotating bowl through a central nozzle. An inlet diversion baffle, located a short distance up in the bowl, prevents any further axial motion of the mixture and diverts it radially (small holes near the center of the baffle, however, allow axial passage of entrained air). The liquids are quickly accelerated to full rotational speed by eight radial vanes that extend the full length of the bowl. The mixed phases separate rapidly in the high centrifugal field (300-500 g), with the heavy (aqueous) phase collecting near the wall and the light (organic) phase collecting near the center. As shown in Figure 2, the organic phase flows inward and over a circular weir in the center of a baffle located at the top of the separating section, and is thrown outward through four, straight-sided, radial ducts to a collector in the stationary casing. At the bowl wall, the aqueous phase passes “under” the circular baffle, then flows inward through the four passages between the radial organic phase ducts into a pneumatically pressured weir section. After overflowing the circular weir in

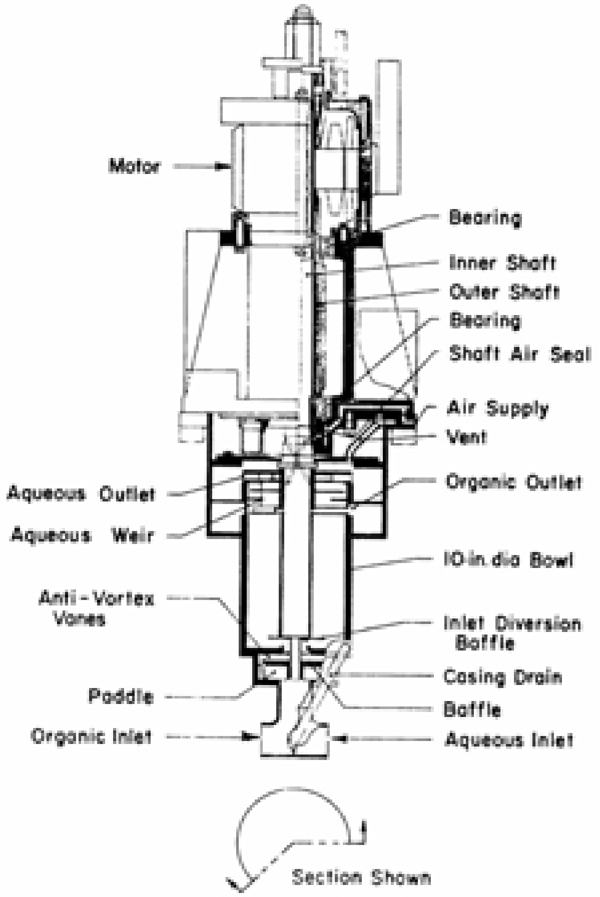

Figure 1. Centrifugal mixer-settler

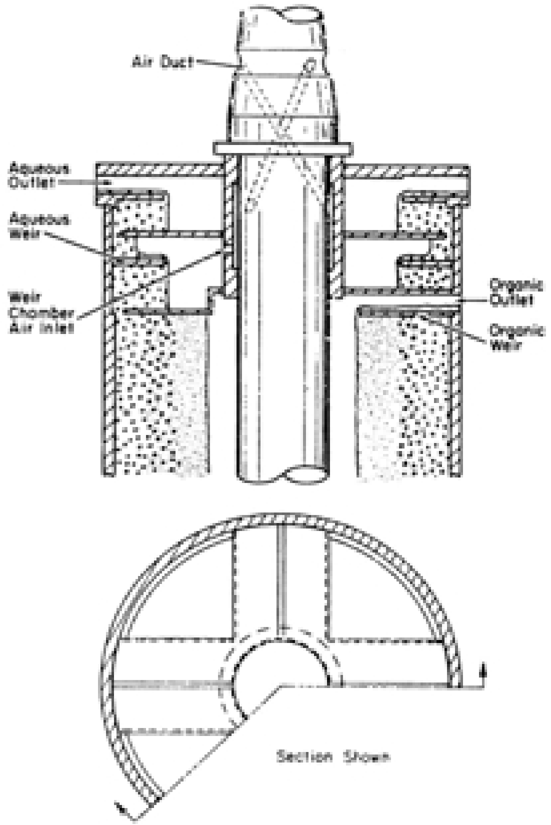

Figure 2. Outlet weir detail

this pressurized chamber, the aqueous phase passes through a circular jack leg seal and is discharged radially into its collector ring. The pressurized weir section allows remote emulsion positioning in each stage. For example, when air pressure is increased, the emulsion moves inward and the surface of liquid in the jack leg moves outward, in compensation.

The two phases flow counter-current between stages. The hold-up of each stage is approximately four gallons (three gallons in the bowl). Compressed air for the weir chamber is transferred via a "Koppers" (Koppers Company trademark) floating bushing air seal from the stationary encasement to ducts in the rotating shaft without significant pressure drop and with a continuous air leakage of less than 1.5 scfm.

Six stages are mounted to form an assembly. Each assembly, with its permanently mounted piping and wiring, and each motor of the assembly, including all bearings and mechanical air seals, are remotely replaceable. Three of the assemblies are piped together remotely to form the 18-stage extraction-scrub bank.

Instrumentation

Each assembly has one control system to supply air at a controlled pressure to the weirs of its six stages. The pressure and total airflow to all six units are recorded in the central control room. Flows to individual stages are indicated by rotometers located just outside the shielding wall. Failure of a seal causes an increase in the total air flow, and the defective seal is then identified as the one with the greatest air consumption, as shown on the individual rotometers.

The power supply to an assembly is divided between two circuits, each serving three stages. The current in each circuit and the difference in circuit between the two circuits are measured and indicated on the central control panel. Motor failure or improper hydraulic operation of a stage causes an unbalanced load and a measurable current difference.

The 3 streams to the 18-stage extraction-scrub bank are the radioactive aqueous feed to a center stage of the bank, an organic extractant to one end of the bank, and an aqueous scrub to the other end of the bank. Successful operation of the process depends upon controlling the flow ratio of the phases to maintain a high uranium concentration in the central stages of the bank. Flow of each stream is controlled by a conventional loop system with a flow measuring device (e.g., a rotometer), a recorder, and a stack controller that delivers an output signal to an air-operated valve or a variable speed pump. The extractant flow is set manually at a value corresponding to the desired processing rate. Flows of the two aqueous streams are controlled relative to this extractant flow by a ratio controller in the controlled loop of each aqueous stream. The ratio of scrub-to-extractant flow is set at a predetermined value and can be changed manually. The ratio of feed to extractant flow is adjusted automatically according to the specific gravity measured in the aqueous phase between Stages 12 and 13. The adjustment keeps the uranium concentration at a specified value. Specific gravity is measured by the differential pressure between two air-purged dip tubes at different depths in a small bypass pot.

An automatic shutdown system stops all feeds and the mixer-settler motors whenever any of the following abnormal conditions occur:

- Low flow of any incoming stream
- High specific gravity in Stage 12
- Low pressure air supply to the weirs
- High differential motor current

Bypass circuits are provided for startup, flushing, and maintenance.

Hydraulic and Mass Transfer Characteristics

Testing the hydraulic capacity of the centrifugal mixer-settlers with radioactive solutions was not feasible because of the inconvenience of any errors. Therefore, the hydraulic and mass transfer characteristics of the numerical 18-stage bank were evaluated at the Semiworks with nonradioactive solutions that simulated those used in the Purex extraction-scrub service. The first test reproduced the low uranium conditions of the waste end of the bank. The flows ranged from rates equivalent to processing 8 tons per day of uranium to 27 tons per day (60 gpm). A maximum processing rate of 14 tons per day was obtainable in the 24-stage bank of pump-mix mixer-settler that the 18-stage centrifugal mixer-settler replaced.

Tests more truly simulating the Purex extraction-scrub conditions were made by introducing a feed of unirradiated natural uranium at Stage 10. The resulting change in relative phase densities required higher air pressures on the weirs for satisfactory performance than did the solution without uranium.

Improper air pressure allows entrainment to pass from one stage to the next, where the entrained phase is separated and returned to the first stage. The increased power required for this internally circulating flow is detected easily and consequently serves as an indicator for improper air pressure.

The mass transfer performance of the 18-stage bank was measured during the tests with unirradiated uranium at processing rates equivalent to 8 to 16 tons per day. The cocurrent stage efficiencies for extraction approached 100%; less than 0.03% of the uranium remained in the waste stream, and less than 0.3% entrainment occurred in each end stream. Each mass transfer test was followed by a Purex 1C-bank operation of the centrifugal mixer-settlers at a rate equivalent to 10 tons per day to back-extract the uranium from the organic phase; the overall mass transfer efficiency was approximately 95%.

Additional Operating Features

Important additional features that apply to the 18-stage unit were previously established with a 4-stage prototype:

1. The hydraulic capacity of centrifugal mixer-settlers is not adversely affected by aqueous-phase acid concentrations as low as 0.01M nitric acid, in contrast to the behavior of pump-mix mixer-settlers, which could not operate below 0.1M nitric acid because of the stability of the resulting emulsion.

2. The method for detecting a motor failure and the procedure for flushing a unit containing an inoperable stage are satisfactory.

3. The air-weir pressures required to control the location of the emulsion can be predicted for any system.

4. The centrifugal mixer-settler performance is not impaired by 5000 ppm of solids in feed solutions. Solids are centrifuged out of solution and accumulate as a thin layer in each stage until the rate of deposition is equal to the rate of re-suspension into the aqueous phase at the underpass baffle. Thereafter, the solids follow the aqueous phase through the bank. In contrast, many of these solids collect at the interface of pump-mix mixer-settlers until the phase separation is greatly slowed.

Plant Performance with Radioactive Solutions

The 18-stage bank of centrifugal mixer-settlers have been highly successful, operating since startup on radioactive feed in 1996. Mechanical reliability has been satisfactory with minimal remote maintenance required on defective air seals or motor bearing failures. Both of these types of failures were remedied by remotely replacing the motor-seal-bearing assembly with a spare assembly. The defective units were decontaminated, repaired, and stored for reuse. One 6-stage module was decontaminated and rewired to replace deteriorated electrical insulation, and another was remotely replaced with a spare because of a crack in the permanently mounted piping. The replaced module was decontaminated, repaired, and stored for reuse. The automatic control system has also performed well. The only change required was damping of the fluctuating signal from the specific gravity probe.

The Purex process has frequently been shut down completely over weekends, or in a few cases, for prolonged scheduled plant outages. The pump-mix mixer-settlers were prepared for shutdown by feeding a “cold” uranium solution for about four hours to purge fission products from the bank and to minimize solvent degradation during the idle period. The total shutdown time was five to six hours. In contrast, the centrifugal mixer-settlers are purged by 15 to 30 minutes of solvent and scrub flow, and shutdown is accomplished in less than an hour. Completely flushing the centrifugal bank, required to obtain product purity when the equipment is used occasionally for processes other than Purex, takes 1 to 2 hours, a marked improvement over 8 to 16 hours involved in completely flushing the pump-mix mixer-settlers. When the previous bank was started after a shutdown, a steady state operation was attained in about 16 hours. As well as it can be determined, the centrifugal bank reaches equilibrium in about 20 minutes.

Decontamination from fission products by the new 18-stage bank has been satisfactory, although somewhat less than that by the old 24-stage bank because of a deficiency of scrub stages. The short residence time and consequent lower exposure of solvent to radiation in the centrifugal mixer-settler has markedly reduced the retention of fission products in the solvent. Gross gamma activity of the solvent, both before and after washing, fell to less than one-fifth of former values for the same feed activities. This reduction permits processing more active feeds. Whereas feeds of 100 curies per liter to the pump-mix mixer-settlers caused excessive fission product retention by the solvent and drastically reduced decontamination through the first cycle, the centrifugal bank has processed feed with up to 250 curies per liter with no adverse affects. No upper limit on feed activity has been established.

The years of experience with the centrifugal mixer-settlers in radioactive service has demonstrated the expected ease of operation and reduction of solvent damage, with good mechanical reliability.

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Biography

Albert A. Kishbaugh received a Bachelor of Science degree in Chemical Engineering from Penn State University in 1952. He was employed by the Du Pont Company in 1952 and was assigned as a Research Engineer at the Knolls Atomic Power Laboratory in Schenectady, New York, while the initial construction of the Savannah River Site was begun. He transferred to the Savannah River Laboratory (SRL) at SRS from 1954 until 1971, at which time he was relocated to the Du Pont Process Section of the Atomic Energy Division in Wilmington, Delaware. While at SRL, he held the positions of research engineer, senior research engineer, and senior research supervisor. While in Wilmington, he was process superintendent in the Process Section from 1972 to 1979 and engineering manager in the Departmental Engineer’s Office from 1980 to 1989, at which time he retired from Du Pont. As engineering manager, he was responsible for coordinating engineering activities between the Du Pont Company Management, SRL, SRS, and the Du Pont Engineering Department. He was employed by Bechtel Savannah River, Inc. (BSRI) from 1989 until 1992, where he held the position of design engineering resident manager at United Engineers and Constructors, Philadelphia, Pennsylvania (1989-1991) and research manager technical representative at Ebasco, Augusta, Georgia (1991-1992).

Development of Pu-239 Processes and Facilities

Edwin N. Moore, Donald A. Orth, Wally B. Sumner, and James A. Purcell

Abstract

During the last several decades, the Savannah River Site (SRS) substantially contributed to the DOE mission success by safely operating and continuously improving the processes and facilities used to make plutonium metal used in nuclear weapons. This paper addresses the noteworthy improvements that contributed to the success of the Pu-239 finishing processes—those that convert plutonium solution produced from recovery of plutonium from irradiated reactor targets into high-quality plutonium metal. The SRS contribution has been to take laboratory-proven processes and convert them into production-proven processes that can safely and efficiently meet DOE mission requirements. The success speaks for itself. *First*, the SRS finishing facilities remain the only Pu-239 production capability in the nation. *Second*, the plutonium efficiency, low waste generation, and excellent product quality from these facilities is unmatched. *Third*, SRS is the only major DOE processing site without a nuclear criticality incident. *Fourth*, although originally built for producing plutonium metal, SRS demonstrated processes that can convert surplus nuclear weapons into oxide for use in fuels. *Fifth*, through SRS equipment and process engineering improvements, the process capacity increased to several times of the original facility design. *Sixth*, the SRS safety record is unmatched.

Overview

The Pu-239 finishing processes convert dilute plutonium solution from solvent extraction processes to a high-purity metal ingot, or button, of plutonium suitable to cast as weapons parts. During Word War II, plutonium-finishing technology developed rapidly from the microgram to the multi-hundred gram scale. The initial finishing strategy, which lasted until several years after World War II, was to ship plutonium nitrate solution, produced at Hanford, to the Los Alamos Site, where it was converted into finished product metal. Cold War expansion, accelerated in the late 1940s, prompted constructing additional finishing capability at Hanford and the Savannah River Site. Included at SRS were additional reactors and new processing facilities, each with a plutonium finishing capability referred to as a B Line.

The SRS canyons and associated B Lines were the first facilities in the world where all the processes to separate plutonium from irradiated reactor elements and convert to high-purity metal buttons were housed in one structure. The initial design of the SRS B Lines was piecemeal, pilot-scale development and lessons learned at other sites including Los Alamos, Hanford, Knolls Atomic Power Laboratory, and Oak Ridge. The challenge for SRS was to modify or adapt the processes and equipment into an integrated process to convert dilute plutonium solution into finished product plutonium metal, meeting the desired start-up capacity and product specifications. Not only was this accomplished, but, through in-plant process improvements based on gained knowledge and understanding of the process chemistry, capacity and efficiency increased significantly. In fact, the process improvements in the three original B Lines (two in F Area and one in H Area, located in the 3rd and 4th levels of the canyon) permitted shutdown of the two F-Area lines to construct the new finishing line while the single H-Area line maintained and exceeded the initial total production output of the original three lines.

The initial processes included:

- Concentration of the plutonium using cation exchange
- Precipitation using hydrogen peroxide
- Drying using warm air
- Conversion to plutonium tetrafluoride using hydrofluorination
- Reduction to plutonium metal using calcium and iodine in an induction-heated furnace

After cooling, the plutonium coalesced in the rounded bottom of the crucible to form the characteristic "button" shape (see Figure 1). A dumping operation separated the button, which was then pickled in nitric acid to remove surface contaminants and packaged for shipping. The resulting plutonium metal product was high quality, with only a small fraction requiring recycle. The slag and crucible separated from the button were packaged for transfer to a recovery process located on the fourth level of Building 221-F.

Figure 1. Plutonium metal button

By the late 1950s, improvements increased throughput reactor fuel fabrication technology (allowing high output per reactor) coupled with increased demand by the Defense Department. To meet this demand, a two-story finishing facility, referred to as JB Line (see Figures 2 and 3), was completed in 1959 on top of the F-Canyon building. The new JB Line used the SRS-developed trifluoride precipitation process and incorporated many facility design improvements from the lessons learned in the original B Lines. In-plant process and equipment improvements continued over the years and allowed JB Line to exceed the government requirements for product output and quality, and to adapt to changing missions. The all-time annual production output record was established in 1983, many times the original output of the initial B Lines. The following sections discuss the substantial improvements (grouped in five areas) made by SRS to the finishing processes and facilities.

Coupling (Cation Exchange)

The thermal evaporation coupling process used at Hanford had several drawbacks, including safety concerns (potential explosive components formed from entrained organic solvent) and process concerns (formation of corrosion products detrimental to the peroxide process and final product purity). The explosion of an evaporator processing uranium nitrate solution at the SRP test facility, TNX, in the early 1950s emphasized the need for an alternative process. Consequently, SRS selected a cation exchange process developed at Oak Ridge, which had several advantages: (1) capability of concentrating the canyon product fiftyfold or more; (2) additional decontamination from uranium, fission products, and anionic impurities; (3) no requirements for high temperature or pressure; (4) no mechanism for formation of explosive compounds from entrained organic solvent; and (5) ability to maintain plutonium in the three (+3) valence state, which proved to be favorable for downstream processing.

The original cation exchange process used at Savannah River Site was developed at ORNL by D.C. Overholt, F. W. Tober, D. A. Orth, and

Figure 2. Building 221-F Canyon with JB-Line

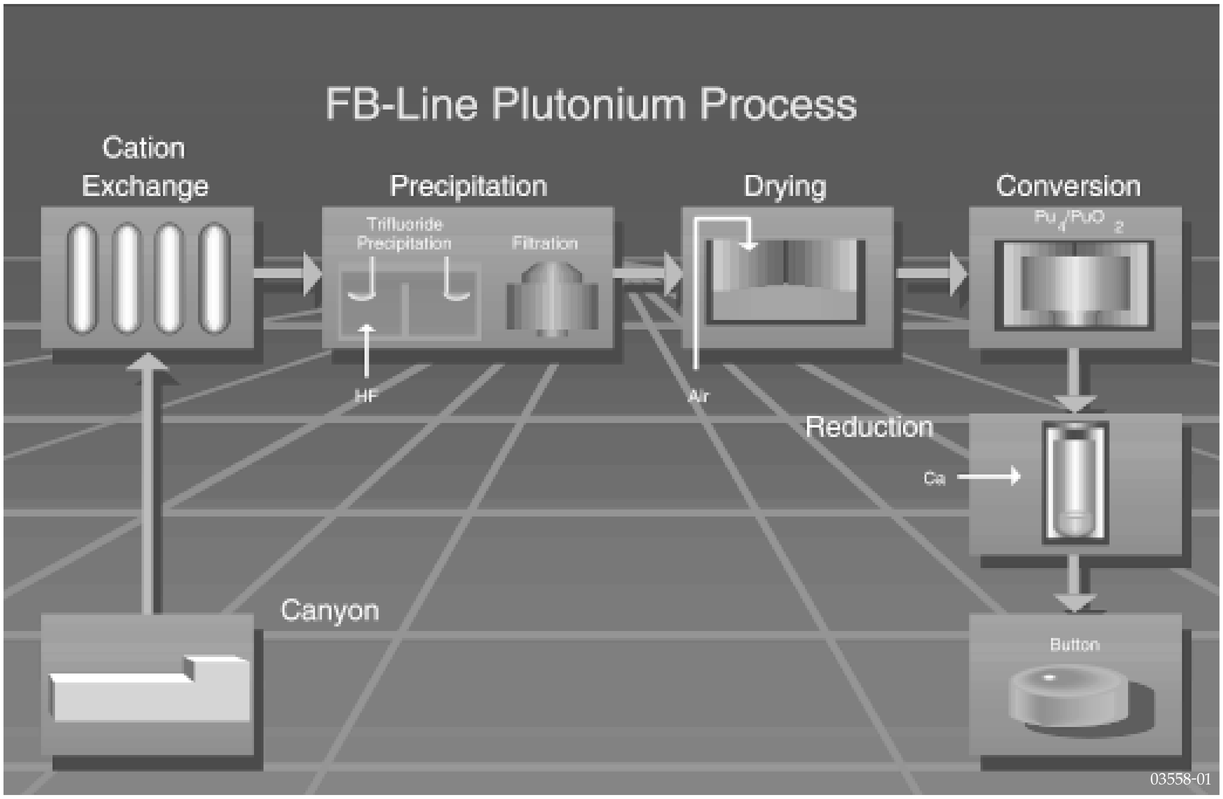

Figure 3. JB Line Finishing Process

others. The process was modified further at Savannah River by Tober and G. W. Burney. Once optimized for plant operation, the basic process has remained unchanged for nearly 50 years and is simple, reliable, and efficient. The resin used is Dowex 50, polystyrene-divinylbenzene beads with exchange sites that are strongly ionized sulfonic acid groups. The plutonium cations, in the three-valence state, exchange with the hydrogen ions during absorption in weak nitric acid solution and are removed during elution with moderate strength acid solution. The optimum resin size, 50-100 mesh, is a compromise between capacity and absorption rates versus bed pressure drop.

The columns are operated on a batch basis with counterflow absorption and elution. The dilute plutonium feed solution, produced by canyon solvent extraction, flows down through the column, until a desired load of plutonium has been absorbed. If necessary, residual uranium and fission products (ruthenium, zirconium, and niobium) that were absorbed with the plutonium are removed using a wash of dilute sulfuric acid. The plutonium is eluted (removed) from the column using an elutriant solution of 5.7 Molar nitric acid and 0.3 Molar sulfamic acid flowing up through the column. A reconditioning wash then flows up through the column to restore the low acid condition for the next absorption cycle. Specific major improvements are as follows:

1. Counterflow operation (i.e., down-flow absorption and upflow elution) was employed to obtain a nearly plutonium-free stripping section at the bottom of the column that produces an effluent waste stream with discard losses, <<0.1%. This approach also reduced the required elution volume and permitted higher concentration in the eluted product.

2. Equipment engineering solutions were employed to increase capacity and improve safety. The use of spring-loaded compressed beds allowed normal bed expansion and contraction (~10%) without deforming or damaging resin beads and without fluidizing the bed, which can reduce column operating efficiency. Increasing the column diameter (10-inch versus 7-inch) and using two short (5-inch versus 1/15-inch) beds in series shortened absorption time while maintaining favorable geometry for nuclear safety control. Using gravity feed systems for all streams fed to the columns eliminated use of a pressurized feed system and associated safety problems. Using ever-open vents alleviates gas buildup in the column, and refrigerated washes, as needed, removed gas pockets, which can reduce bed absorption efficiency.

3. Optimization of column load and elutriant compositions precluded auto-catalytic oxidation of the plutonium, which can produce rapid gas generation and pressurization of the column. The use of sulfamic acid in the elutriant provided a stable holding reductant in the moderately high acid concentration.

Precipitation

Peroxide Precipitation

The initial SRS process employed peroxide precipitation, developed at LANL. Unfortunately, the LANL flowsheet was demonstrated on concentrated plutonium solutions; hence, substantial effort was required to adapt this flowsheet and equipment for the lower concentration SRS feed solutions. The peroxide process was selected because of

- Its excellent decontamination for cationic impurities
- Its minimum introduction of foreign material
- Its readily filterable precipitate (if the chemistry is adequately controlled)
- The excellent reactivity of its precipitate in the subsequent hydofluorination process

After adjusting the plutonium nitrate feed solution to +3 valence and 4.7 molar nitric acid, the solution is fed to a cooled vessel where concentrated hydrogen peroxide is slowly added to form plutonium peroxide. After a digestion period, the plutonium peroxide is separated from the filtrate by filtration. To aid filtration, a small amount of sulfate is added as a coagulant for colloidal particles, and is incorporated in the crystal structure. Cooling is necessary to minimize catalytic decomposition of the hydrogen peroxide from trace impurities (e.g., iron) in the feed. The initial process involved washing the cake with dilute hydrogen peroxide, then with ethyl alcohol to aid drying, which is performed using dry, heated air. Any residual peroxide present in the filtrate is destroyed by heating the solution at 50 degrees C, then at 90 degrees C prior to recycle.

If conditions were not properly controlled, long filtrations and high plutonium losses would result. Considering that the original precipitator vessel was simply a larger (to allow for more dilute SRS feed) version of the one used at LANL, with the same agitator as used at LANL, the original poor performance is not surprising. Process improvements are as follows:

1. Overall process yields of 99.5% and reduced filtration times were achieved by optimizing precipitator vessel design, reagent addition rate, and agitator speed, eliminating zones of localized low acid conditions, which favored the formation of fine, difficult-to-filter particles. These changes produced a precipitate six times as dense that filtered six times as fast and eliminated sulfate additions, which substantially reduced in subsequent high-temperature roasting and hydrofluorination time cycles, otherwise required to remove sulfate and its interference with reduction to metal.

2. Air-drying improvement eliminated alcohol (a fire hazard) washes of precipitate that gave temperature and pressure excursions in the hydrofluorination furnaces.

3. By 1957, the cycle time was reduced to almost half that of the original system. Other improvements include a larger batch size, made possible by higher nuclear safety limits following neutron multiplication studies, and increased precipitant concentrations resulting from using 50% versus 30% hydrogen peroxide. In addition, improvements in filter boat design and materials of construction (stainless steel versus platinum) enhanced filter life and improved filtration and drying.

Trifluoride Precipitation

In the late 1950s, the trifluoride process was developed at SRS and installed as the precipitation process in the JB Line. The trifluoride process was selected because it eliminated stability and corrosion concerns with the hydrogen peroxide and hydrofluorination processes, and because it produced a more readily dried precipitate and with less plutonium loss to the filtrate. However, the trifluoride process proved less capable of decontamination of impurities and resulted in higher operator radiation exposure. Corrosion issues with using concentrated hydrofluoric acid in precipitation offset the elimination of corrosion issues associated with HF gas. The process proved very sensitive to upsets; hence, careful control of chemistry and equipment engineering proved crucial. Nevertheless, this process has proven reliable and has served SRS effectively for several decades.

Before precipitation, the plutonium nitrate solution is adjusted to +3 valence and 4 molar nitric acid. Precipitation takes place in a two-stage precipitator. Hydrofluoric acid and plutonium nitrate solution are simultaneously added to the first stage under conditions that favor crystal growth. The resultant slurry overflows to a second-stage precipitation vessel where conditions are controlled by adding more hydrofluoric acid to reduce the solubility of the plutonium fluoride and to complete the precipitation. The resultant slurry is then filtered to collect a plutonium trifluoride cake in a filter boat. The cake is washed with dilute hydrofluoric acid and then dried using warm (70-100 degrees C) air for 3-4 hours, resulting in a cake with 2-3% moisture content. Process improvements are as follows:

1. The use of geometrically favorable vessels, with higher criticality limits, permitted an increase in the precipitation batch size from a few hundred grams of plutonium in the original B Line to over a kilogram in the JB Line.

2. Process development identified the ideal conditions for filtration, which required control of the free fluoride concentration (F^-) as measured by a ratio of HNO_3/HF in the first stage precipitator around 7 to 1. A higher ratio promotes too much crystal growth, resulting in excessive solids accumulation on the vessel walls, which necessitates more frequent equipment clean-out. A lower ratio results in fine particles that are difficult to filter. A ratio of 3 or less is necessary in the second stage to reduce losses.

3. Improved vessel and agitator design eliminated dead zones and provided adequate suspension without breaking up particles. Improved materials of construction (Teflon™ and polypropylene) reduced corrosion, improved performance, and extended life of filters and agitators. Further, ascorbic acid proved an effective agent in maintaining the plutonium valence in the +3 state, which is necessary to avoid solids buildup and filtration problems.

Oxalate Precipitation

During the 1960s, plutonium oxide was produced to fabricate into fuels to use in the SRS reactors to make heavy isotopes and in the Hanford pilot breeder reactor. The oxalate process was adapted for use at SRS by John Porter and Albert Symonds. In this process, the plutonium solution is first adjusted to an acid concentration of less than 1.5 M (preferably less than 1 M), and the plutonium valence is reduced to +3 using sulfamic and ascorbic acid. The plutonium is then precipitated in a single-stage vessel by the slow addition of 1 M oxalic acid. After a brief period of digestion, the plutonium oxalate is collected on a filter boat and washed to remove excess acid. The "pool table green" filter cake is then dried to remove free moisture, heated to remove excess waters of hydration, and then converted to plutonium oxide in a oxygen atmosphere at around 700 degrees C. The resultant oxide has particle properties that are ideal for fuel fabrication use. Further, the properties of the final oxide can be controlled as required by controlling the precipitation parameters.

New Precipitators

A replacement trifluoride precipitator system (see Figure 4) was installed and started up on May 13, 1987. Replacement was necessary because of the high radiation exposure from plutonium trifluoride (300 times greater than from plutonium metal) and extensive corrosion from HF, which severely damaged the equipment and glovebox containment. The new system incorporated the following improvements.

1. Remotely operable, electrically operable valves and instruments allowed operation of the system from a distributed control system. These enhancements substantially reduced exposure and improved process control. The new design resulted in a 75% reduction in radiation exposure and process downtime.

2. The new design virtually eliminated removing glovebox windows for maintenance. All equipment, with the exception of large vessels, was designed to be maintained through gloves. This improvement greatly reduced the Pu assimilation risk in maintenance using temporary plastic containment enclosures.

3. Design improvements enhanced control over system chemistry resulting in uniform, consistent operation. New instruments and valves with remote, chemically resistant sensors and seals allowed remote process

Figure 4. Modern control room for replacement precipitators

operation. An electrical connector allowed quick disconnect of electrical lines and glovebox penetrations without exposing sensitive electronics to a corrosive acid environment. In addition, advances in construction materials (e.g., Kynar) and improved vessel and agitator design allowed more uniform operation.

Conversion/Reduction

Hydrofluorination/Reduction of Plutonium Peroxide

The original SRS process dried plutonium peroxide and then converted it to plutonium tetrafluoride. This process, called hydrofluorination, took place in a furnace at 600 degrees in anhydrous hydrogen fluoride (HF) gas. The resulting tetrafluoride was mixed with metallic calcium and an iodine booster and placed in a magnesia crucible, which is nested within magnesia sand within a steel pressure chamber. This vessel is heated in an induction coil until an initiation of a highly exothermic reaction, which reduces the plutonium to metal. After cooling and disassembly, the plutonium metal button was separated from slag and crucible, which contains residual plutonium, which requires recovery. Improvements are as follows:

1. During the first few years of operation, SRS substantially improved this process, doubling of the batch size and reducing the time cycle fourfold. This large increase in capacity concurrently improved the precipitator, drying system, and eliminated sulfate, as previously discussed, coupled with improved furnace cooling and sealing. The net result was that hydrofluorination was no longer a limiting step.

2. The larger batch sizes eliminated iodine in 1957. Using iodine caused molten reaction products to fuse through the ceramic liner of the reduction pressure vessel, making button dumping very difficult. Iodine in the reduction slag also gave problems in subsequent plutonium recovery processing.

3. Dramatic improvements in supporting systems were also achieved. Low reduction yields in the early years of operation were

presumed due to deviation from procedures; hence, technical engineers were assigned to each shift to oversee operation. However, upon investigation, it was found that reduction yields are extremely sensitive to glovebox humidity, a problem not encountered at LANL or Hanford. Replacing degraded reagents and fixing the air drying system, which was not cycling properly, restored high yields. Other improvements included redesigned induction coils, which greatly reduced coil failures, and ultrasonic pickling, improving button quality.

Conversion/Reduction of Plutonium Trifluoride

In the initial JB-Line process, plutonium trifluoride precipitate was dried and roasted in an argon atmosphere at high temperatures to remove any volatile impurities. The roasted trifluoride was then mixed with calcium and an iodine "booster" for reduction to plutonium metal. Equipment engineering improvements, including improved criticality safety design and a remotely operable dry-processing line, tripled batch size and reduced radiation exposure to the operators. Although this new process eliminated the need to use gaseous HF, it introduced other problems, such as the reintroduction of iodine additions. Specific improvements include the following:

1. Thermodynamic analysis and test indicated that the addition of oxygen to the roasting step would convert the trifluoride to a mixture of plutonium tetrafluoride and plutonium oxide, which could be reduced without the addition of an iodine booster. Adoption of this procedure resulted in reduction yields that were about 5% lower than those using iodine. This lower yield was considered an acceptable price to pay to eliminate iodine and coincident reduction in recovery time cycle, corrosion, and offgas concern.
2. In recent upgrade projects, new roasting/conversion furnaces have been installed with improved shielding, offgas handling, and process controls.

Recovery and Special Recovery

The B Lines, both old and new, had facilities to recover plutonium from solid and liquid residues resulted from the main finishing processes. This precluded accumulating and indefinite storage of these residues. These residues consisted primarily of

- Slag and crucibles from the reduction process
- Floor sweepings from the dry cabinets
- Scrap plutonium metal (reject buttons or sample turnings)
- Button pickling solutions
- JB-Line precipitator flushes

The original recovery facilities were located on the fourth level of the 221-F Canyon Building. After JB Line start-up, a replacement recovery facility was installed in JB Line. The original facility employed various recovery processes.

In the original B Lines, slag and crucibles and/or cabinet sweepings were recovered in a dissolver vessel containing concentrated nitric acid, aluminum nitrate, and water. Aluminum nitrate is added to partially complex the free fluoride normally present from dissolving solids. Free fluoride is necessary, and added as needed, during the dissolution step to dissolve entrained particles of plutonium metal or any high-fired oxide that may be present. The dissolver is purged with nitrogen during the initial phase of dissolution to safely remove hydrogen produced from the dissolution of excess calcium metal in the slag. After the dissolution cycle at the simmering temperature, the solution is slightly cooled, and aluminum nitrate is added to completely complex the fluoride and to minimize downstream corrosion. The solution is digested at the elevated temperature to coagulate silica, introduced as an impurity in the crucible. After cooling, the solution is filtered to remove the coagulated silica and other insoluble materials. The filtered solution is purified by solvent extraction or anion exchange. JB-Line precipitator flushes and pickling solutions are filtered and added with the dissolver solutions for purification. The original recovery process used PUREX solvent extraction in small mixer-settler banks inside wet process cabinets. Process improvements are as follows:

1. Dissolving slag and crucibles with iodine released copious quantities of iodine, corroding the dissolver offgas system. Eliminating iodine coupled with hot caustic flushes to clean the filters substantially improved recovery operations and reduced the cycle time by over a factor of two.

2. Sulfamic acid was accidently discovered to be an excellent dissolution media for plutonium metal in 1957, which greatly reduced the cycle time and corrosion encountered with boiling nitric/hydrofluoric acid dissolution. SRS effectively used this process for years to recover on- and off-site metal. The dissolution takes place at slightly elevated (about 45°C) temperatures, producing hydrogen gas, and is some cases, pyrophoric sludge. In the mid 1980s, discovery indicated adding a small amount of hydrofluoric acid to the sulfamic acid dissolving solution eliminated pyrophoric sludge formation.

3. The solvent extraction system was replaced by a simpler, more-compact anion exchange process. The feed solutions for anion exchange are adjusted to a total nitrate concentration of about 8–9 M to form an anionic complex that is readily absorbed by anion resin (e.g., Dowex 21K). The column is eluted with dilute nitric acid to produce a plutonium product solution, which is diluted to a safe concentration and transferred to the canyon.

4. As early process and equipment difficulties were overcome, the throughput capacity and versatility of the recovery facilities became sufficient to not only process the SRS residues, but also to process various residues from other sites including Los Alamos, Hanford, Argonne, and Rocky Flats. Many types of non-routine residues were processed including: Lampre fuel (tantalum-clad plutonium); ZPPR fuel containing oxides of plutonium, uranium, and molybdenum; high chloride solutions; plutonium/aluminum alloy; plutonium containing neptunium; test residues containing fused mixtures of plutonium, oralloy, and extraneous materials; plutonium sludge recovered from the F-Canyon air tunnel; and plutonium residue from offgas filters. SRS became the "can do" plutonium recovery facility for much of the AEC/DOE complex.

5. A New Special Recovery Facility was built and ready for startup in 1991; however, the reduced demand for plutonium for defense eliminated this facility. It was constructed to process 2 MT (metric tons) of plutonium oxide per year, and was being modified, when shutdown, for handling up to 3 MT/year of either oxide or metal feed. Processes were developed to separate plutonium from various pit types, to eliminate concerns with pyrophoric sludge that plagued earlier metal dissolving efforts, and to reduce hydrogen generation. The facility was designed for remote control room operation similar to that described for the New Precipitators.

6. In recent years, concerns over hydrogen generation with existing recovery flowsheet for calcium-laden reduction residues led to developing direct canyon dissolving processes, which are currently in use.

Facility Engineering Improvements

Old “B” Lines

Many design features of the first-generation facilities built at LANL were not suitable for full-scale production facilities (e.g., many operations took place in open face hoods). Consequently, many operators assimilated small amounts of plutonium. The situation was further complicated in that surfaces within the facility were difficult to decontaminate. The initial SRS finishing lines, called “B” lines were just a step up from the laboratory stage. For example, some reagents used in the process were added using calibrated pails. Worse, the equipment was not designed for easy maintenance (e.g., only six gloveports were installed in each line for routine operations). In the original facilities, the process equipment was located in two types of negative pressure enclosures—wet cabinets surrounding aqueous processes and dry cabinets surrounding powder-handling processes. The wet cabinets used valve extension handles operate valves without using gloveports and employed air in-leakage for contamination control. The dry cabinets were hermetically sealed and supplied with dry air through inlet and outlet HEPA filters. Although a mechanical conveyor system was provided in the dry cabinets to remotely move materials among stations, it proved unreliable; hence, material movements were done manually. Although some design improvements (more gloves for maintenance, new central vacuum system, and localized radiation shielding) were incorporated, maintenance frequently required removing cabinet panels, which required constructing plastic huts for contamination control. Nuclear criticality safety in the original button lines was maintained with small batch sizes, about 300 grams plutonium, and with vessel size controls.

New JB Line

The new JB Line had many facility and equipment engineering improvements to correct lessons learned in the original B Lines. Within the first year of operation, these improvements provided the capacity to handle in 15 shifts of operation the output from the canyon in 21 shifts of operation. The new facility used vessels with neutron-scattering geometry and other controls for nuclear criticality safety that permitted processing batches seven times larger than in the original lines. For the dry process, duplicate stations for every process step existed within wing gloveboxes along a main, mechanical conveyor line to prevent failure of a single station from shutting down the entire process line. Where possible, gloveports accessed equipment for maintenance. Room partitions compartmentalized work areas for contamination control. The gloveboxes were designed for routine operations on one side and maintenance on the opposite side, allowing simultaneous maintenance and processing and helped to control contamination. Improved construction materials (e.g., plastic tanks and coated agitators) improved equipment life in the corrosive hydrofluoric and nitric acid glovebox atmosphere. In addition, at- and in-line plutonium solution monitoring systems increased throughput by providing results in one-half hour, and in some cases real-time, that had previously taken 4 hours from the Laboratory. Improved assay instruments detected mis-operation (filter failure, over-batching, etc.), which substantially improved safety and process efficiency.

Restoration Program and Recent Improvements

During the 1980s, extensive facility restoration activities were performed, like replacing some process operations (i.e., precipitators, conversions furnaces, vent and vacuum systems, and the neutralization system) and restoring facility piping systems, cabinet panel seals, and ventilation systems. During the last decade, the facility underwent extensive outside review and improvement in integrated safety management and conduct of operations. Materials control, accountability, fire protection, materials characterization, waste management, security, and safeguards were upgraded. Installing a bagless transfer system enabled metals packaging in welded containers, eliminating storing plastic bags for containment. Additional improvements to package plutonium to meet new storage standards are under development. The current facility mission is to characterize and stabilize onsite and selected off-site surplus plutonium inventories.

The Future of Finishing Processes

Continuing use of finishing technologies is essential to convert surplus plutonium metal to oxide suitable for Mixed Oxide Fuel, as currently planned. Many of the SRS developed technologies and engineering approaches may prove useful in this purpose. For example, SRS developed processes to dissolve the plutonium metal or oxide away from other materials and to produce a high quality oxide with minimal radiation exposure and waste generation. Most the engineering technology to perform process operations remotely has been developed. Evidencing the efficiency of the SRS process is the fact that there is less plutonium in transuranic solid waste at SRS than at any other major DOE site. The successful improvement over the years to the finishing processes is directly due to the SRS determination to understand and apply equipment-engineering solutions to improve processes.

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Development of Chemical Processes and Equipment to Recover Curium-244 and Californium-252

Robert M. Harbour, Clark H. Ice,
William H. Hale, and John T. Lowe

Abstract

In the mid 1960s, ^{244}Cm and ^{252}Cf were produced at the Savannah River Site (SRS) from ^{239}Pu starting material in nuclear reactor irradiations designed for successive neutron capture. Both solvent extraction and ion exchange processes and equipment were developed at the Savannah River Laboratory (SRL) to separate and purify curium and californium from irradiated plutonium-aluminum fuel tubes. High-pressure ion exchange proved more suitable than a solvent extraction process with tertiary amines ("Tramex") for the final stages of purification. A combination of high-pressure displacement and elution development cation exchange chromatographic processes was able to separate and purify 100 g quantities of ^{244}Cm and milligram quantities of ^{252}Cf. Curium-244 is suitable as an isotopic power source for remote applications, and ^{252}Cf is an intense neutron source for medical and industrial applications.

Introduction

In the mid 1960s, kilogram quantities of ^{244}Cm and milligram quantities of ^{252}Cf were produced by irradiating ^{239}Pu in a Savannah River Plant (SRP) reactor. The ^{239}Pu was converted to ^{244}Cm, and, in turn, ^{252}Cf and other transplutonium elements were converted by successive neutron captures and beta decays during irradiation of the aluminum-plutonium fuel tubes:

$$\text{Pu} \rightarrow \text{Am} \rightarrow \text{Cm} \rightarrow \text{Bk} \rightarrow \text{Cf} \rightarrow \text{Es} \rightarrow \text{Fm}$$

The SRP program for producing transplutonium elements in several stages is depicted in Figure 1. In the first irradiation, ^{239}Pu captured neutrons to produce heavier isotopes. The resulting mixture of plutonium isotopes was recovered (Curium I Campaign) and fabricated into aluminum alloy targets for a second irradiation in which a portion of these plutonium isotopes was converted to ^{242}Pu, ^{243}Am, and ^{244}Cm. The bulk of the ^{244}Cm from targets of this second irradiation (Curium II Campaign) was separated and purified for evaluation as an isotopic power source for remote applications such as space satellites. The ^{242}Pu and ^{243}Am were recovered and re-irradiated along with some of the ^{244}Cm to produce ^{252}Cf and other transplutonium elements such as berkelium, einsteinium, and fermium. The spontaneous fission mode of radioactive decay of ^{252}Cf makes it an ideal portable neutron generator for medical and industrial applications.

SRL developed a pilot-scale two-stage solvent extraction process using (1) tributyl phosphate (TBP) in the first stage to recover plutonium and separate a crude actinide-lanthanide mixture (Curium I, II Campaigns) and (2) tertiary amines (designated Tramex and conceived at Oak Ridge National Laboratory) in the second stage to purify curium and americium from rare-earth fission products (Californium I Campaign). Californium was contained in the curium fraction.

The Tramex solvent extraction process consists of three cycles of extraction with a tertiary amine to separate the trivalent actinides from the lanthanides. This process requires high-concentration chloride solutions and proved difficult to control. A 'skunk works' effort was begun at SRL to demonstrate the technical feasibility of an alternative approach based on ion-exchange chromatography. In 1968, Nat Stetson (manager, SR Operations Office), Lom

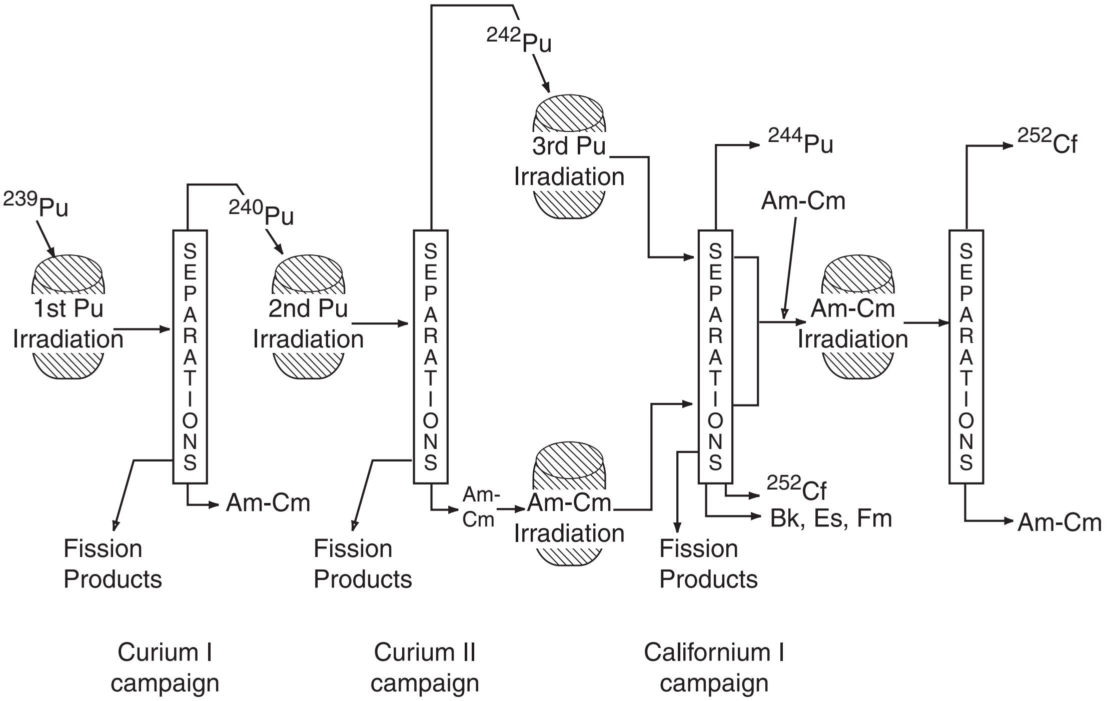

Figure 1. Outline of the transplutonium elements production program at Savannah River Plant

Squires (manager, AED), Clark Ice (director, SRL), and Bill Hale and John Lowe (research chemists) met in Washington, D.C., with Glenn Seaborg (chairman of the Atomic Energy Commission) to review initial positive results of a new approach, high-pressure cation exchange chromatography for separation and purification of curium and californium. Armed with Seaborg's endorsement, SRL launched a major effort to develop a large-scale high-pressure cation exchange chromatographic process. What followed was an excellent team effort between the AEC and Du Pont with leadership and support from Frank Baranowski, who headed up the AEC Division of Production, to successfully produce and purify large quantities of ^{244}Cm and ^{252}Cf.

Advantages of High-Pressure Cation Exchange

Conventional low-pressure cation exchange was an effective method both for separating transplutonium actinides from lanthanides and for separating adjacent transplutonium elements. However, low-pressure cation exchange proved ineffective for large quantities of highly radioactive materials such as curium, americium, and californium because of the relatively long residence time on the column. The localizing heating and radiation due to radioactive decay energy degrades the resin material, and radiolytic gases generated by radiolysis of the solution physically disrupt the resin bed. Pressurizing the cation exchange system reduced the residence time of these highly radioactive feed materials and dissolved the radiolytic gases, suggesting that separation and purification of curium and californium were theoretically possible.

Pressurized cation exchange also has advantages over solvent extraction processes. The pressurized cation exchange process requires less accurate control of concentration and flow rates than the solvent extraction process. In addition, cation exchange requires only stainless steel equipment, whereas tantalum or Zircaloy-2 is required for the chloride solutions in the alternative solvent extraction methods.

In developing a high-pressure cation exchange process suitable for curium and californium, optimal operating parameters needed to be developed for resin particle size, elutriant flow rates, column length, pressure levels, elutriant composition and concentration, feed concentrations, column temperature, and lanthanide/ actinide elution sequence. In high-pressure cation exchange, finely divided resin particles (5-10 or 30-60 mm) are used instead of 100-200 mm or larger resin particles in conventional cation exchange. Elutriant flow rates from 1-50 ml/min-cm^2 have been obtained with positive displacement pumps pressurizing the feed and elutriant solutions up to 2500 psig. A pressure limit is finally reached where a further increase in pressure does not result in increased flow due to resin compaction.

With small-particle resin, equilibration between solution and resin is rapidly attained and results in potentially better separations than in conventional cation exchange. Rapid equilibration for small-particle resin results from decreased diffusion distances in both the resin particles and in the aqueous film surrounding the particle. The use of small-particle resin improves kinetics and gives a more uniform flow, which allows fast flow rates for the loading and elution steps. These high flow rates drastically reduce the time necessary to attain a given separation. The decreased residence time of highly radioactive feed materials in the resin reduces radiation damage to the resin, especially during the loading step. Also, localized heating due to radioactive decay energy being deposited in the system is reduced.

To achieve the fast flow rates possible with small-particle resin, elutriant and feed solutions are pressurized, which has the additional advantage of dissolving the radiolytic gases produced. In highly radioactive solutions, radiolytic gases are generated by radiolysis of the solution primarily by alpha particles and fission fragments from decay of transplutonium actinides.

Radiolytic gases can cause resin bed disruptions, which may result in irregular flow and channeling. Some gassing may occur at the bottom of the column, but the high pressure keeps bubble size small and removes the gas immediately. When the columns are operated at elevated temperatures, solution viscosity is reduced, and the pressure drop is lower for a given flow rate.

Because cation exchange resin shows little selectivity among lanthanides and actinides, a chelating agent must be used to separate cations of the individual elements into separate bands. Two types of chromatographic development were used in the actinide separations: displacement development and elution development. Displacement development is superior for large-scale separations. Rates of band movement and product concentrations in displacement development are approximately tenfold greater than those in elution development.

Displacement Development Chromatography

Displacement development involves the following steps:

- The resin is loaded with a cation (the barrier ion) that has less affinity for the resin than the ions to be separated.
- A mixture of the ions to be separated is loaded onto the column.
- A chelating agent having different chemical complex stability with each of the cations to be separated is pumped through the column.

The cations of each element in the actinide-lanthanide mixture form separate bonds and are eluted with the elements forming the strongest chemical complex with the chelating agent coming off ahead of the others. There is a binary zone, or overlap region, between each band of pure component because the column is not at equilibrium.

In 1968-69, a high-pressure displacement development cation exchange chromatographic process was developed at SRL to replace the Tramex solvent extraction process for separation of large quantities of transplutonium actinides. As much as 250 mg of ^{252}Cf and 190 grams of ^{244}Cm were separated in a single batch from americium and lanthanide fission products using Zn^{2+} as the barrier ion and 0.05M diethylenetriamine pentaacetic acid (DTPA) at pH 6.0 as the chelating agent. The system consisted of four 4-foot columns in series with decreasing diameters from 4 inches to 1 inch. Each column was loaded with 25-55-µm-particle-size “Dowex” 50W cation exchange resin (synthetic polymer containing sulfonic acid groups) at 70°C. Successively smaller columns were used because the length of the overlap zone is independent of diameter, but the amount of material in this zone decreases with diameter. A high pressure (900 psig) pump forced the DTPA solution through the columns at a flow rate of 16 ml/min-cm^2.

The results of a typical displacement development separation, which took 8-16 hours, are shown in Figure 2. Californium-252 elutes ahead of ^{244}Cm and is not shown in this figure. Three kg of ^{244}Cm, 1.3 kg of ^{243}Am, and 8 mg of ^{252}Cf were separated using this system.

Elution Development Chromatography

Elution development consists of the following steps. The cation resin is conditioned, and a mixture of the ions to be separated is loaded onto the column to form a narrow band near the top of the column. Complexing agents that form chemical bonds of different stability with the various ions to be separated are used to achieve selective elution from the resin. There exists a competition between the resin and the complexing agent for each ion, and if the column is run close to equilibrium conditions, each ion will be exchanged between the resin

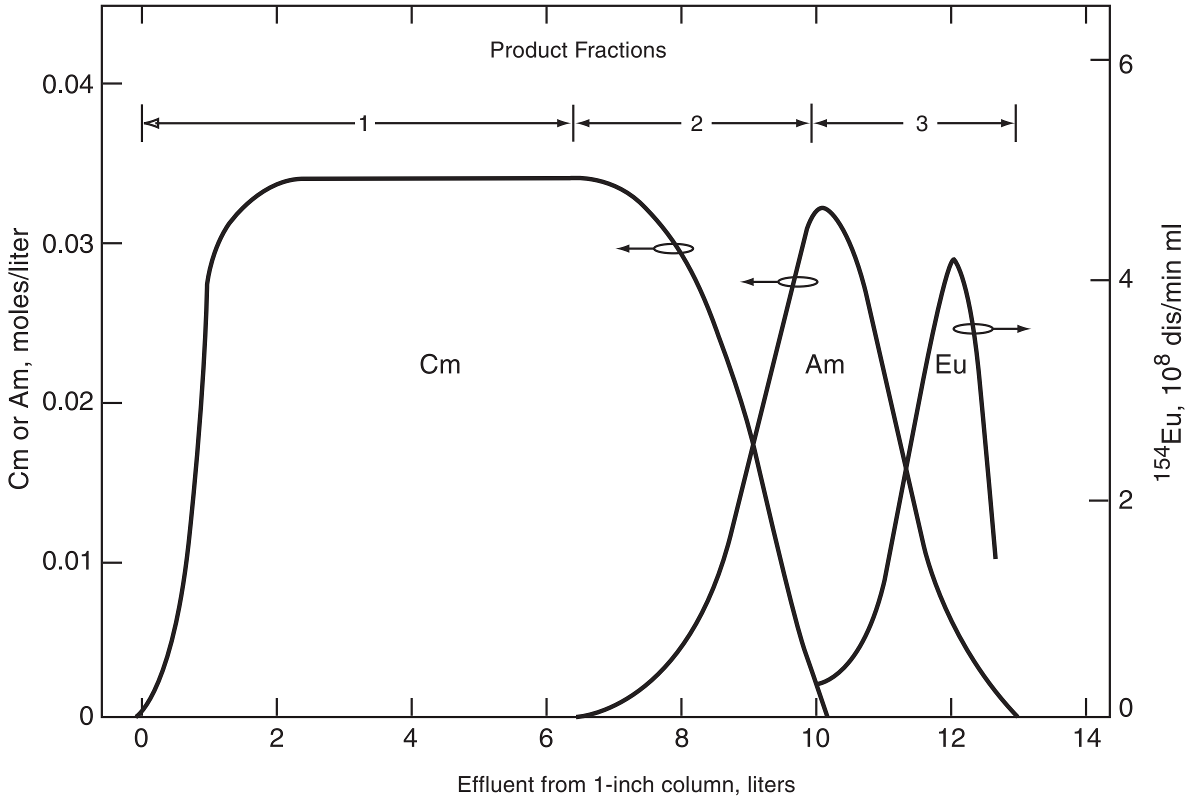

Figure 2. Typical displacement development diagram

and complex form many times as it moves down the column. As the various absorption bands move down the column, their spatial separations increase until finally the ion from the lowest band appears in the effluent from the column.

In 1968-69, a high-pressure elution development cation exchange chromatographic process was also developed at SRL for separation and purification of ^{252}Cf containing fractions from both Tramex and DTPA displacement development processes. Milligram quantities of ^{252}Cf, submilligram amounts of ^{249}Bk, and subnanogram amounts of ^{253}Es and ^{254}Fm were separated from multigram quantities of ^{244}Cm and ^{243}Am using 0.4M alphahydroxy isobutyrate (AHIB) partially neutralized with NH_4OH to a pH of 3.4 as the elutriant. The pH of this AHIB solution was increased stepwise during the elution as shown in Figure 3 to effect a sharper separation. The system consisted of one 120-cm-long, 5-cm-diameter column loaded with 20-40-µm-diameter Dowex 50W cation exchange resin at 75°C. A high-pressure (700 psig) pump forced the AHIB solution through the column at a flow rate of 8 ml/min-cm^2. The results of a typical elution development separation, which took 5 hours, is shown in Figure 3. Over a two-year period, 3.5 mg of ^{252}Cf were purified from Tramex solvent extraction feed material, and 5.5 mg of ^{252}Cf were purified from DTPA displacement development cation exchange feed material.

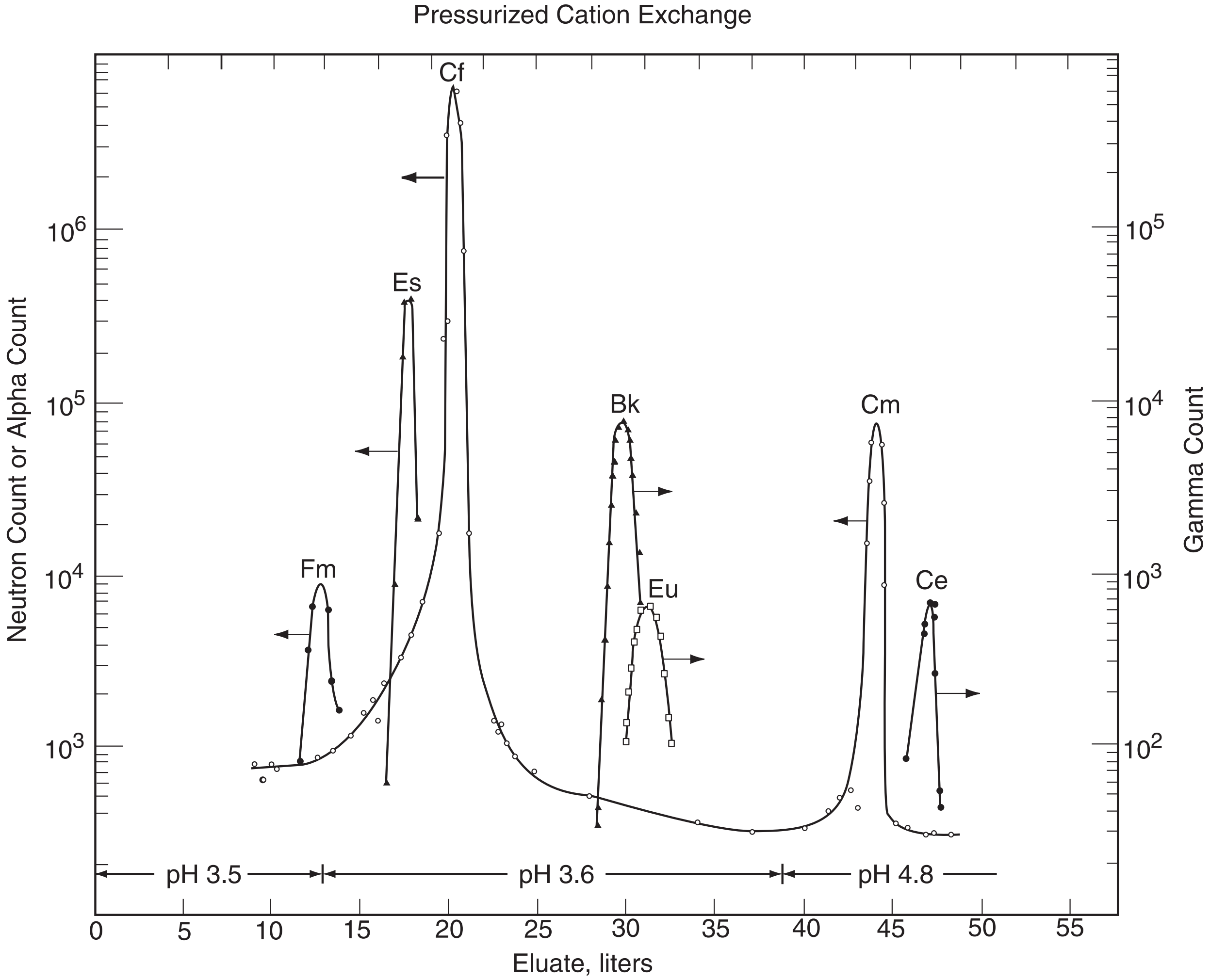

Figure 3. Typical elution development diagram

Uses of ^{252}Cf and ^{244}Cm

Californium-252 is a unique source of neutrons because it yields a highly concentrated and reliable neutron spectrum from a very small assembly. During the past 40 years, ^{252}Cf has been applied with great success to cancer therapy; neutron radiography of objects ranging from delicate flowers to massive objects, such as entire aircraft; startup sources for nuclear reactors; elemental analysis of coal, nuclear fuel, explosives, and the human body; and many other beneficial uses. For many of these applications, ^{252}Cf is unique and irreplaceable. From 1965 to 1995, 8 g of ^{252}Cf have been produced, 3.6 g were shipped, and 600 mg were sold. A Market Evaluation Program sponsored by the Department of Energy determined that at that volume of usage, the large-quantity production capability of ^{252}Cf at SRL was not required. Therefore, the Oak Ridge research facility continues to meet present and projected near-term ^{252}Cf requirements at a price of $50/mg. Californium-252 is also produced in Russia.

Curium-244 was made available for development and demonstration of SNAP generators and of isotopic power fuels for deep space probes, but these uses have not come to fruition. Small quantities have been used as a gamma source, but its primary use has been as target material for producing ^{252}Cf. Curium-244 has a specific heat generation rate of 2.8 w/g and decays with an 18-year half-life. Curium-244 sells for $185 per milligram and can also be purchased from Oak Ridge National Laboratory.

Site Impact

The Site impact of the Curium and Californium Campaigns included several reactor cycles at SRP to produce sufficient quantities of ^{244}Cm and ^{252}Cf. Each reactor run was followed by work in the separation areas to do initial solvent extraction separations. The development of the TBP and Tramex solvent extraction separation processes for this application was done at SRL. The novel high-pressure displacement and elution development cation exchange chromotographic separation and purification processes were also developed at SRL.

Biographies

Robert M. Harbour

Robert Harbour worked at SRL from 1968 to 1973, first as a research scientist, then as a research supervisor. During the balance of his 25-year Du Pont career, he worked in Petrochemicals and Imaging Systems Departments in a variety of research, manufacturing, marketing, and business management positions both domestically and overseas. From 1984 to 1985, he succeeded Frank Kruesi as Director, Technology Division for AED.

Post-Du Pont, Mr. Harbour managed an early-stage venture capital firm in Sydney, Australia, and currently has a management consulting firm in Santa Fe, New Mexico, focussed on assisting high-growth, technology-oriented companies.

Mr. Harbour graduated from Macalester College, St. Paul, Minnesota, in 1964 with a B.A. degree in chemistry, and from the University of Missouri, Columbia, Missouri, in 1968 with a Ph.D. in physical chemistry. He did post-doctoral work at Oak Ridge National Laboratory, Oak Ridge, Tennessee, before joining Du Pont at Savannah River Laboratory (SRL).

Clark H. Ice

Clark Ice worked at the Hanford site from 1947 to 1950 as an analytical chemist before arriving at SRP's Laboratories Section in 1952. In 1954, he moved to SRL as senior supervisor in the Analytical Chemistry Division. He subsequently held positions as research manager, Chemistry Division; director, Separations Chemistry and Engineering Section; and assistant director, SRL, before becoming director, SRL.

Following his retirement from Du Pont, Mr. Ice worked for General Public Utilities (GPU) from 1979 to 1989 as a member of the Three Mile Island (TMI) 2 Technical Advisory Group and the TMI 2 General Office Review Board.

Mr. Ice graduated with a B.S. from Central State Teacher's College, Edmond, Oklahoma, in 1938. In 1941, he received a Master's Degree in chemistry and in 1952 a Ph.D. in biochemistry, both from the University of Oklahoma, Norman, Oklahoma.

William H. Hale

William Hale worked at SRL from 1966 to 1976, first as a research chemist, then as a research supervisor and research manager. During the balance of his 28-year Du Pont career, he held a variety of management positions in Petrochemicals and Information Systems Departments before moving to Conoco in 1988 as Vice-President for Conoco Information Systems. Following his retirement in 1993, he was Director of the National Supercomputing Research Centre in Singapore during 1994-1996. He currently works as a consultant with Du Pont Safety Resources and with E. G. Mahler & Associates.

Mr. Hale graduated from The University of Texas in 1963 with a B.S. in chemical engineering and from the University of California-Berkeley in 1966 with a Ph.D. in inorganic chemistry. He attended the Stanford Executive Program in 1990.

John T. Lowe

John Lowe worked at the Savannah River Laboratory from 1965 to 1971, first as a research chemist, then as a research supervisor. After a series of management assignments in the Petrochemicals Department, he returned to Savannah River in 1983, sequentially as program manager, laboratory director, and plant manager. In 1991, he returned to the Petrochemicals Department as director of research and development.

Following his retirement from Du Pont in 1993, he was director of the process industry practices initiative, a consortium of 30 major owner and contractor firms developing a common set of engineering standards for the process industry. He is currently associate director of the Construction Industry Institute, and is a consultant with E. G. Mahler & Associates.

Mr. Lowe graduated from Georgia Institute of Technology in 1962 with a B. S. in chemistry, and from the University of Illinois with a Ph.D. in physical chemistry in 1965.

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Development and Performance of Processes and Equipment to Recover Neptunium-237 and Plutonium-238

Harold J. Groh, W. Lee Poe, and John A. Porter

Abstract

During the late 1950s, the Atomic Energy Commission (AEC) requested the Savannah River Site (SRS) to begin as soon as possible to produce plutonium-238 for use as a heat source for thermoelectric generators to power satellites. Thus, SRS joined the United States space race with the former Soviet Union, as well as continuing as a key facility in the race to produce materials for thermonuclear weapons.

To accomplish this new task, a massive interdisciplinary effort was required by all of the technical, engineering, maintenance, and administrative staff. New chemical and metallurgical processes were developed and installed in ingenious new equipment in the existing production buildings, with minimum interference with the processes operating to produce weapons materials.

Neptunium-237, the precursor isotope to the production of plutonium-238, was recovered from the existing processes, where it had been produced as a byproduct in the reactor irradiation of uranium. The purified neptunium-237 was fabricated into targets, which were neutron irradiated in SRS reactors to produce plutonium-238. The plutonium-238 was separated from neptunium and fission products and processed to plutonium oxide. The unconverted neptunium-237 was purified, processed to neptunium oxide, and used in additional reactor targets. Plutonium-238 oxide was fabricated into dense fuel forms at SRS and at other sites for use in thermoelectric generators.

Plutonium-238 produced at SRS has powered about two dozen spacecraft launched by NASA, including Transit, Viking, Voyager, Galileo, and Cassini.

Introduction

The Savannah River Site (SRS) began operation in the early 1950s to produce nuclear materials for weapons for the national security effort. The United States was in a weapons race with the former Soviet Union to produce thermonuclear weapons. There was great pressure on the staff of SRS to produce as rapidly as possible the tritium and weapons-grade plutonium (consisting predominantly of the fissionable plutonium-239 isotope) needed for nuclear weapons.

In 1957, the Soviet Union launched its first satellite, Sputnik, and began a race to space with the United States. In 1958, the United States launched its first satellite, Explorer I. In 1960, the Soviets put the first man into space. In 1961, President Kennedy started the National Aeronautics and Space Administration (NASA) program to put a man on the moon. In the late 1950s, the Atomic Energy Commission gave SRS the mission to produce a new reactor product, plutonium-238. This lighter isotope of plutonium, which had been made in only experimental quantities up to that time, was needed as a heat source for thermoelectric generators to power satellites. (Plutonium-238 has one less neutron per atom than its heavier isotope, plutonium-239.) Thus, SRS became an important part of the space race as well as continuing to be a key facility in the weapons race. The first plutonium-238 was made at SRS in 1961.

All of the work during these early years was conducted under strict rules of secrecy to conceal technical details and production quantities. In fact, in the early program, secrecy was so tight that code names were used to conceal the identities of the isotopes neptunium-237 and plutonium-238.

Plutonium-238 has several characteristics that make it almost unique for fueling a radioisotope thermoelectric generator (RTG). It decays by emitting alpha particles with accompanying evolution of heat that can be readily converted to electricity. Gamma radiation from plutonium-238 is predominantly of low energy, requiring little shielding, which reduces the weight required for space applications. The relatively long half-life (about 88 years) makes it suitable for deep space missions, where sunlight is too weak to generate much electricity from solar cells.

Plutonium-238 fueled RTGs have provided power for about two dozen spacecraft launched by NASA, including:

1961	The first RTG delivered 2.7 watts of electrical power to a Navy Transit navigational satellite.
1969-72	Apollo astronauts left RTGs on the moon to power experiments.
1972	Pioneer spacecraft flew by Jupiter and Saturn.
1975	Viking spacecraft landed an instrumented vehicle powered by an RTG on the surface of Mars.
1977	Voyager 1 and 2 spacecraft flew past Jupiter and Saturn (three 150-watt electrical RTGs on each spacecraft).
1989	Galileo flyby of Jupiter (two RTGs, 285 watts electrical each) was launched. Exploration of Jupiter and its moons is underway.
1997	Cassini probe to explore Saturn (three RTGs, 285 watts electrical each) was launched.
1999	Galileo flew by Jupiter's innermost moon, Io.

Over the next 10 years, NASA is planning several more missions that are expected to use plutonium-238 for electric power.

Several challenges had to be faced in the development of production technology for plutonium-238.

- Processes and equipment had to be developed that were compatible with the existing plant processes. Time was not available to construct new production buildings. The new processes would have to be incorporated with minimum interference into the facilities that were already operating to separate weapons-grade plutonium from uranium, and these new processes must not slow the production of weapons.
- There was no available supply of the precursor element, neptunium-237. It would have to be recovered and purified first before the production of plutonium-238 could begin.
- The radioactivity of plutonium-238 is about 250 times that of plutonium-239, so radiation damage to process materials, such as organic ion exchange resins, was expected to cause problems. In addition, there were concerns about the adequacy of containment of plutonium-238 in conventional, filtered glove boxes.

Technical Basis for Plutonium-238 Production

The precursor isotope, neptunium-237, is produced when uranium is irradiated in nuclear reactors. The nuclear physics reactions are the following (Groh 1970)

$$^{238}\mathrm{U}\,(n,\,2n)\;^{237}\mathrm{U} \xrightarrow[6.7d]{\beta^{-}} {}^{237}\mathrm{Np} \qquad (1)$$

$$^{235}\mathrm{U}\,(n,\,\gamma)\;^{236}\mathrm{U}\,(n,\,\gamma)\;^{237}\mathrm{U} \xrightarrow[6.7d]{\beta^{-}} {}^{237}\mathrm{Np} \qquad (2)$$

The reactions illustrate the production of neptunium-237 as a byproduct from the irradiation of either natural uranium, Equation (1), or uranium enriched in the uranium-235 isotope, Equation (2). This byproduct had been produced at SRS since startup but had been lost to the radioactive waste streams during chemical processing of the irradiated fuel elements. It was necessary to develop modifications to the existing chemical separations processes to isolate the neptunium and to purify it from uranium, plutonium and fission products. In SRS reactors, neptunium production from enriched uranium fuels was the predominant route, particularly after the practice of recycling the irradiated enriched uranium was begun to conserve the intermediate isotope, uranium-236.

Once neptunium-237 is recovered and purified, it is converted to plutonium-238 by returning it to the reactor in target elements, where the reaction shown in Equation (3) takes place:

$$^{237}\text{Np}\,(\text{n}, \gamma)\,^{238}\text{Np} \xrightarrow[2.1d]{\beta^{-}} {}^{238}\text{Pu} \qquad (3)$$

Short exposures of the neptunium targets in the reactors limit the production of the higher isotopes of plutonium. Typical SRS plutonium-238 product had an isotopic composition of approximately 81% plutonium-238, 15% plutonium-239, 2.9% plutonium-240, and lesser amounts of plutonium-241 and plutonium-242. Unavoidably, some of the neptunium and plutonium are converted to fission products during the irradiation, and these must be removed during the chemical processing.

In the late 1950s, when work was begun at SRS on processes to recover neptunium-237 and plutonium-238, a large body of information already existed in the literature on the chemistry of neptunium and plutonium. But it was necessary quickly to augment that data in some areas and to apply it to practical separations processes that could be operated in the SRS plants. The byproduct neptunium was recovered from the solvent extraction processes operating in the two canyon buildings by a combination of modifications to the solvent extraction processes to divert the neptunium and ion exchange to recover and purify it. An anion exchange technology was chosen to process the recovered neptunium after it was re-irradiated to form plutonium-238. One of the most important chemical properties of neptunium and plutonium in solution is the ability of these elements to exist in several oxidation states or valences. This is an important tool for the separation of these elements where their behavior is controlled primarily by valence adjustment by oxidizing and reducing chemicals.

The varied oxidation states and ionic species of neptunium and plutonium that can exist under normal conditions are shown in Table 1. Uranium, by contrast, normally exists only in the VI oxidation state in aqueous solutions, as the UO_2^{2+} ion.

The solvent extraction processes are based upon the preferential extraction from nitrate solution of the actinide elements into tri-n-butyl phosphate (TBP) dissolved in dodecane to separate them from contaminating elements such as aluminum and the fission products. The order of extraction into TBP from nitrate solutions is: U(VI) > Np(VI)   Pu(IV) > Np(IV)   Pu(VI) >> Np(V), Pu(III). Thus, U(VI) and Np(IV) can be separated readily from inextractable Pu(III); and U(VI) can be separated from Np(V). Aluminum and most of the fission products are inextractable in TBP. The solvent extraction

Table 1. Oxidation States of Neptunium and Plutonium

Oxidation State	III	IV	V	VI
Neptunium Ions		Np^{4+}	NpO_2^{+}	NpO_2^{2+}
Plutonium Ions	Pu^{3+}	Pu^{4+}	PuO_2^{+}	PuO_2^{2+}

process operated in the F-Area canyon building separated weapons-grade plutonium from natural uranium or depleted uranium (uranium from which the uranium-235 isotope had been partially removed). This was called the Purex Process. The H-Area Canyon process, called the HM Process, recovered enriched uranium from irradiated fuel elements.

The anion exchange process is based upon the fact that Np(IV) and Pu(IV) form anionic nitrate complexes of the type $Np(NO_3)_6^{2-}$ that are strongly absorbed from concentrated nitrate solutions by strong-base anion exchange resin, such as Dowex 1-X4. The other oxidation states of neptunium and plutonium are very weakly absorbed, as are most fission product species and common metallic cations. Thus Np(IV) and Pu(IV) can be effectively separated from uranium and fission products by anion exchange, and Np(IV) can be separated from Pu(III).

Description of Processes

Recovery of Neptunium-237 from Purex and HM Processes

In the Purex Process, one modification was needed to ensure that essentially all of the neptunium was directed to the high-level waste streams from which it was to be recovered. Nitrite was added to the extraction section of the first solvent extraction contactor in which uranium and plutonium are extracted into TBP to maintain the neptunium in the (V) oxidation state and thus minimize loss to other waste streams. (This contactor was originally a mixer-settler, but later was replaced by centrifugal contactors.) The high-level waste streams were subsequently combined and concentrated by evaporation in preparation to recover neptunium by anion exchange with new modular equipment installed in F Canyon. The concentrated waste was adjusted to 8M nitric acid and treated with ferrous sulfamate and hydrazine nitrate to produce Np(IV), which forms a strong anionic nitrate complex, and was fed to an agitated (stirred) anion exchange column to absorb the neptunium. Most of any plutonium-239 that was present was also absorbed as it too forms a strong anionic nitrate complex. The agitated resin column, which will be described in a later section, was necessary to accommodate solids that are typically present in concentrated wastes. After the feed step, the column was washed with 8M nitric acid to remove impurities, and then the neptunium and plutonium were eluted with 0.35M nitric acid.

The solution from the first bed was processed further to separate neptunium and plutonium on a fixed bed of anion exchange resin as follows. Solution acidity was adjusted to 8M, and valence adjustment was repeated using ferrous sulfamate and hydrazine stabilizer, followed by heating to 50°C to oxidize Pu(III) to Pu(IV). The solution was fed to the resin bed, which absorbed the neptunium and plutonium. The bed was washed with 8M nitric acid to further remove impurities, and the plutonium was removed by washing with 5.5M nitric acid containing ferrous sulfamate and hydrazine to reduce plutonium to the (III) state. The recovered plutonium-239 was returned to the Purex second plutonium cycle. The neptunium was eluted with 0.35M nitric acid, processed by another cycle of anion exchange for further purification, followed by a cycle of cation exchange if necessary for thorium removal.

In the HM Process, operating conditions were modified to isolate and purify neptunium entirely by solvent extraction in the existing equipment. The dissolver feed was treated with ferrous sulfamate to maintain the Np(IV) valence state so that neptunium was extracted along with uranium in the 1A mixer-settler and partitioned from uranium in the 1B mixer-settler. Further decontamination of neptunium was accomplished using the Purex flowsheet in second plutonium cycle equipment that was not otherwise being used. The neptunium product solution was concentrated by evaporation in preparation for further processing in HB Line (a lightly shielded, glovebox facility in Building 221-H).

Isolation of neptunium from the HM Process continued for the life of the plutonium-238 program, but was discontinued from the Purex Process after an adequate inventory was accumulated.

The nitric acid solutions of neptunium from the Purex and the HM Processes were transferred to the HB Line for conversion to neptunium dioxide, NpO_2, which could be fabricated into targets for neutron irradiation to produce plutonium-238. In HB Line, neptunium was further purified and concentrated, if needed, by anion exchange. After anion exchange, the concentration of nitric acid was adjusted within the range of 1-2M, and the valence of the neptunium was adjusted to the (IV) state with ascorbic acid and hydrazine inhibitor at about 50°C. Neptunium(IV) oxalate was precipitated at the same temperature by the addition of 1M oxalic acid solution. After digestion to aid crystal growth, the slurry was cooled to room temperature and filtered and air-dried. Neptunium dioxide was produced from the neptunium oxalate by heating in air to a final temperature of about 550°C (Porter 1964).

Recovery of Neptunium-237 and Plutonium-238 from Irradiated Targets

Processing of irradiated targets to recover plutonium-238 product and residual neptunium-237 was initially performed on a small scale in the High-Level Caves and B-Wing facilities of the Savannah River Laboratory. For the continuing larger scale operations, the very high radioactivity levels associated with the neptunium and the plutonium required installation of new plant equipment in the heavily shielded separations processing areas. Thus, unique modular units called “frames” (to be described later in this paper) were installed by novel techniques in H Canyon to process the irradiated targets. HB Line was also modified to provide facilities to convert the purified plutonium-238 product and the recovered neptunium-237 to the final oxide forms.

A schematic diagram of the neptunium target recovery process is shown in Figure 1. The irradiated NpO_2-aluminum targets, containing unconverted neptunium-237, product plutonium-238, and fission products, were cooled at least 45 days to allow decay of short-lived fission products and then dissolved in boiling 10M nitric acid containing small amounts of mercuric ions and fluoride ions as catalysts. Dissolution was slow, requiring up to 48 hours for about 85% dissolution. Any undissolved target material was carried over and dissolved with the subsequent batch of targets. The dissolver solution contained small quantities of solids, primarily silica, which was removed by filtration prior to further processing.

The plutonium-238 product and residual neptunium-237 were separated from aluminum, fission products, other impurities, and from each other by anion exchange processing. The resin columns were normally operated as fixed beds, but could be operated as agitated beds when needed for regeneration or replacement of resin. In the first cycle of anion exchange, neptunium and plutonium were both adjusted to the (IV) valence state in 8M nitric acid by treatment with ferrous sulfamate and hydrazine, followed by heating to 50°C. The neptunium and plutonium were absorbed on the resin column to separate them from aluminum, fission products, and other cationic impurities. After washing with 8M nitric acid for decontamination, the neptunium and plutonium were eluted with 0.35M nitric acid. In the second cycle of anion exchange, the solution was adjusted to 8M nitric acid, and neptunium and plutonium were again both adjusted to the (IV) valence state and absorbed on the resin. After washing with 8M nitric acid for further decontamination, the plutonium was separated from the neptunium by washing the column with 5.5M nitric acid containing ferrous sulfamate and hydrazine to reduce the plutonium to the (III) valence state. The neptunium was then eluted with 0.35M nitric acid. The separated neptunium and plutonium were processed by one or more additional cycles of

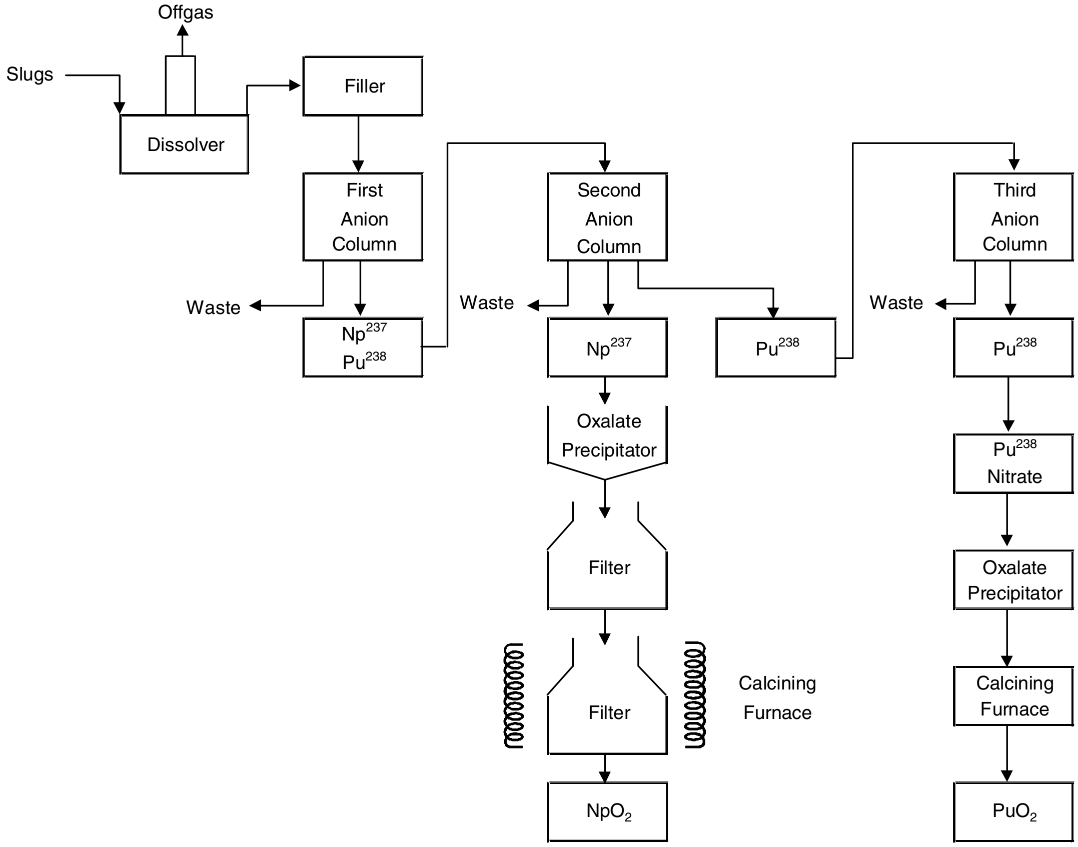

Figure 1. ^{238}Pu recovery from irradiated ^{237}Np at Savannah River

anion exchange as needed to obtain the desired purity. The purified neptunium solution was transferred to HB Line for further processing to produce NpO_2 for recycle. The purified plutonium solution was further processed as described below.

Conversion of Plutonium-238 to Oxide for Use in Heat Sources

In the early phases of the program, plutonium-238 was isolated as a dilute nitric acid solution that was shipped to Mound Laboratory in Miamisburg, Ohio, for additional processing and fabrication into heat sources. Later, to improve shipping safety, the plutonium was processed further in the HB Line to the solid oxide form either for shipment to another site or for onsite fabrication of heat sources.

In the HB Line, the plutonium solution from H Canyon was processed by anion exchange, if required, for decontamination and concentration. Then, the nitric acid concentration was adjusted within the range of 1-2M and plutonium valence was adjusted to (IV) by adding ascorbic acid and hydrazine nitrate and heating to 50°C. Plutonium(IV) oxalate was precipitated at the same temperature by the addition of lM oxalic acid solution. After digestion to aid crystal growth, the slurry was cooled to room temperature and filtered and air-dried. Plutonium dioxide was produced by heating the plutonium oxalate in air to a final temperature of about 550°C.

An innovative technique was used at times in HB Line to reduce the rather high emission rate of neutrons from $^{238}PuO_2$. Neutron emission results from spontaneous fission of plutonium-

238 and also from (α,n) reactions with impurities and with the small quantities of ^{17}O and ^{18}O isotopes that occur in natural oxygen, which is mostly ^{16}O. Research at Mound Laboratory and at Savannah River Laboratory showed that the ^{17}O and ^{18}O content of $^{238}PuO_2$ can be reduced to very low values by isotopic exchange with ^{16}O. This can be readily accomplished by heating the plutonium oxide in the presence of oxygen gas or water vapor that is highly depleted in ^{17}O and ^{18}O. In HB Line, this was implemented by flowing isotopically pure $^{16}O_2$ over the oxide at a temperature of a few hundred degrees centigrade. Neutron emission rates were reduced to near the value expected from spontaneous fission and impurities, and resulted in lower radiation exposure of personnel.

Some of the $^{238}PuO_2$ produced at SRP was shipped to other sites for production of heat sources, but a large number of heat sources was also produced onsite in a new facility installed in 1978 in Building 235-F. D. T. Rankin describes the production of heat sources at SRP in another paper in this symposium proceedings.

Radiation Effects of Plutonium-238

In the chemical processes, the high radiation level of plutonium-238 was dealt with in several ways. Perhaps the most serious problem was radiation damage to the organic ion exchange resins. This was controlled by choosing resins, such as Permutit SK, that were more resistant to radiation, by limiting the exposure times of the resin to plutonium-238, by agitating the resin beds between cycles to redistribute the resin particles, by maintaining continuous flow of process solution through resin beds loaded with plutonium-238, and by periodic remote replacement of the resin. In solutions, radiolysis products from plutonium-238 alpha particles interfered with the stabilization of the desired oxidation states of plutonium and neptunium. This was controlled by the addition of reducing agents and stabilizers such as hydrazine. A combination of ferrous sulfamate and hydrazine proved to be most effective for maintaining the lower oxidation states of neptunium and plutonium while minimizing undesirable side effects such as gas formation.

Conventional gloveboxes provided adequate containment of plutonium-238 in the final steps of processing. The boxes were constructed of stainless steel and operated at subatmospheric pressure with several stages of high efficiency filtration for the box offgas. Local shielding for gamma and neutron radiation, including leaded gloves, protected the workers from radiation exposure.

Description of Production Equipment

Unitized Frame Concept

The recovery of neptunium-237 from the Purex and HM processes and the recovery of plutonium-238 from irradiated neptunium targets involve gamma radiation levels that are equivalent to those encountered in processing spent nuclear fuels; therefore, these processes were installed within the shielded space of the Savannah River canyons. The term canyon refers to the heavily shielded buildings (Buildings 221-F and 221-H) that house and supply services to process equipment for recovering plutonium and uranium from spent reactor fuels.

The canyons at Savannah River provide a module 10 feet square by 17 feet high for each vessel having standard services (i.e., inlet and outlet piping, steam, cooling water, electricity, sampling, and instruments). Each building section has four modules, isolated by a low curb to contain liquid spills or leaks, and has a sump and permanently installed transfer jet to move spilled liquids or leakage to a rework system. Transfers between vessels in the canyon are made on a pipe rack in which the pipe is remotely removable. A direct jumper can make transfers between adjacent vessels.

As discussed above, ion exchange was selected as a key process for recovery, separation, and decontamination of neptunium-237 and plutonium-238. The ion exchange vessels and flows of solutions to and from them are much smaller than those used in the normal solvent extraction processes for which the canyons were designed. Ion exchange processes had never before been installed and operated in a facility that was remotely maintained. Since experimental work performed by the Savannah River Laboratory showed that it would be necessary, from time to time, to replace the ion exchange resins in the canyon process vessels, a technique was developed to transfer them to and from the canyon as slurry (Bebbington 1990).

The recovery of neptunium from depleted uranium required installation of four ion exchange columns in F Canyon. The recovery of neptunium-237 and plutonium-238 from irradiated neptunium targets required five ion exchange columns and a dissolver in H Canyon (Poe 1964). Unitized frame design was adopted for these units to minimize canyon space requirements. The frame concept means installing a number of small equipment pieces in a steel frame that is installed and removed by the normal canyon crane. The specialized equipment was designed by adapting the small-scale equipment to remote operation in the canyon. After building a complete prototype unit to test the feasibility of the concept, three units were constructed and used many years in the canyons. Figure 2 is a photograph of one these frames. A fourth unit was installed in F Canyon for initial recovery of neptunium. This unit consisted of a single ion exchange column and its support tankage.

Placing several equipment pieces in a frame in a single canyon module is economical of canyon space but places a heavy burden on the services available in that module. Pipe and electrical connections were made to the frame rather than to individual pieces of equipment. Services were piped to the equipment as part of the permanent frame structure. This efficiently used existing services, but additional services were required, primarily for pneumatic liquid level and specific gravity instrumentation for several vessels in each frame. Bundles of up to 6 stainless steel tubes (1/4-inch o.d.) were drawn through the 3-inch pipes embedded in the canyon shielding walls. Demonstration of this concept had great utility in adapting existing facilities to alternate processes. No significant problems were encountered in installing these lines, and their useful life has been equivalent to the life of embedded piping.

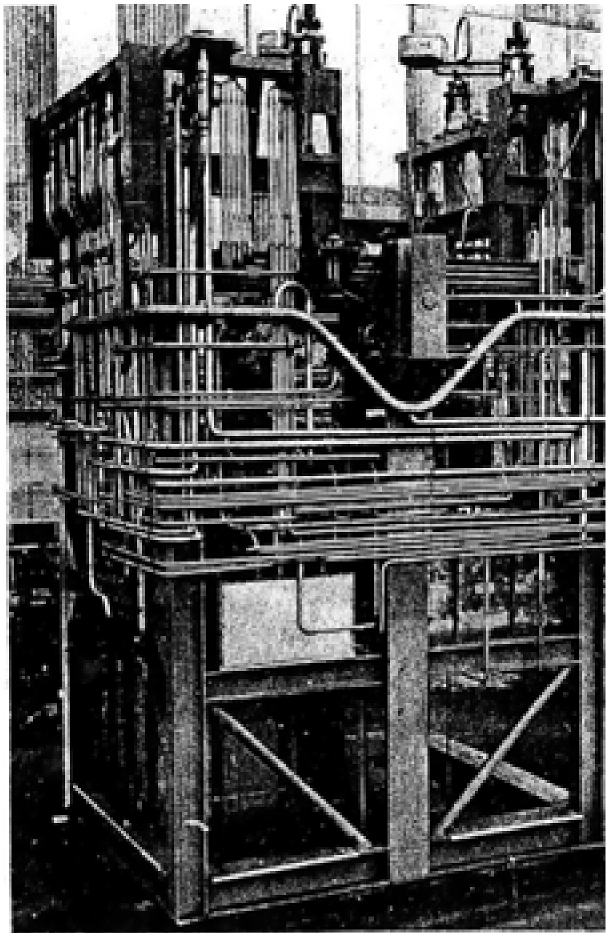

Figure 2. Np and ^{238}Pu frame (Complete frame without ion exchange columns. Frame as installed remotely with Canyon crane.)

Ion Exchange Columns

The ion exchange columns that were built, tested, and used in these operations were either

12- or 24-inch diameter and normally contained 25 or 100 liters of resin. They normally were used as conventional settled bed ion exchange columns but had the capability of use as agitated ion exchange bed columns. One unit was used as an agitated bed column. All operations, including resin replacement, were performed remotely without mechanical valves. Resin was charged and discharged as free-flowing slurry in a solution having a specific gravity close to that of the resin. Two weirs determine flow paths; air pressure is applied to either or both to direct flow properly for feed, wash, elution, or resin removal. Figure 3 shows an isometric diagram of one such ion exchange column.

Dowex 1 and Permutit SK anion exchange resins were used. Dowex 1 resin provides higher decontamination from fission products, but Permutit SK resin is more stable under alpha radiation. Dowex 1-X4 (4% nominal cross linkage) was used in 40 to 60 mesh range particle size. Selected use of smaller particle resins, some with cross linkages as small as 2%,

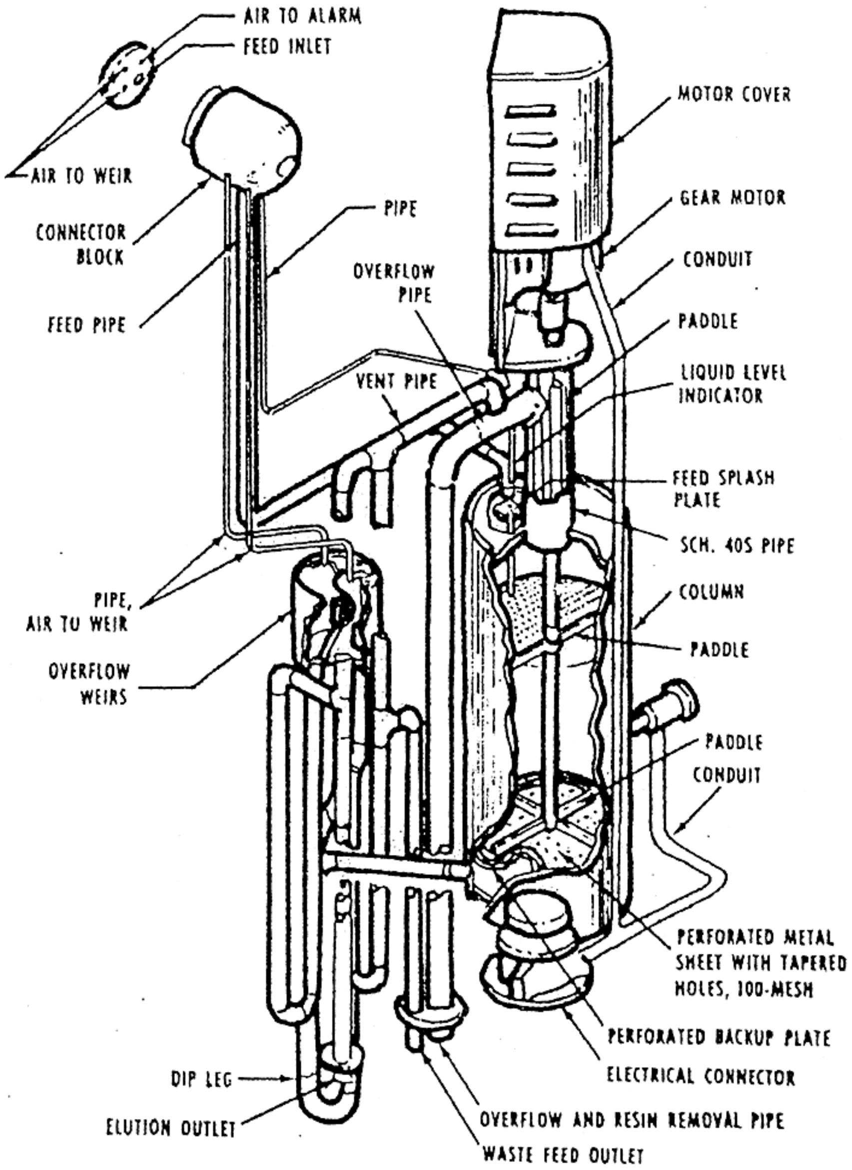

Figure 3. Ion exchange column (for installation on frame shown in Figure 2)

was used where fast kinetic absorption was required, such as in the ion exchange column used for separation of plutonium-238 from neptunium-237. Solution flow through the resin was determined by the resin particle size and gas generated from alpha radiolysis.

Performance of Production Processes

The losses of either neptunium-237 or plutonium-238 in the H-Canyon frame operation used for irradiated neptunium targets averaged 2.7% for the three neptunium processing columns and 4.9% for the four plutonium processing columns. This material was discarded to High-Level Waste for the first two years of operation, and then the frame waste was diverted to a new frame waste column and a further 95 % of this was recovered. High fission product decontamination was accomplished; the limiting fission products were the radionuclides zirconium and niobium. This process gave a decontamination factor (curies in the feed divided by curies in the product) of 100,000 for neptunium and 1,800,000 for plutonium. The second ion column was used to separate the plutonium from the neptunium; this was one of the most crucial activities in the target processing because cross-contamination of one product left in the other product would be lost to waste. Using the Dowex 1-X2 resins, removal of 97 to 99% of the plutonium from the neptunium could be achieved. (This ~1 to ~3% of the plutonium lost to the neptunium is included in the 4.9% waste loss described above.)

HB-Line Facilities for Conversion of Neptunium and Plutonium to Oxides

Decontaminated neptunium-237 and plutonium-238 products from the H-Canyon Frames were converted to NpO_2 and PuO_2 in HB Line, after an initial period of liquid shipments of plutonium-238 nitrate to Mound Laboratory. Initially these operations were performed in facilities designed for weapons plutonium finishing. These facilities had been made obsolete when H-Canyon operations were changed from Purex (weapons-grade plutonium) to processing highly enriched uranium in the late 1950s. Neptunium-237 and plutonium-238 finishing operations were performed in HB Line gloveboxes. The plutonium-238 operations were performed in a new small glovebox line, and the neptunium was finished in the old B-Line cabinets. After several years of operation in this manner, it was decided to construct a new HB Line on top of Building 221-H designed for these operations. The new line had three subparts: one for neptunium, one for plutonium-238, and the third for recovery of neptunium and plutonium from scrap. This new facility was located over Sections 2 through 6 of Building 221-H. It consists of the new fifth and sixth level of that building.

The new neptunium oxide line was constructed over Sections 4 and 5 of the 221-H Canyon. The oxide line consists of two glovebox lines constructed with adjacent operating and maintenance rooms to minimize spread of contamination. The line decontaminated and converted neptunium nitrate to neptunium oxide. Processes employed include ion exchange, precipitation, calcination to oxide, and packaging of the neptunium for shipment to Building 235-F for fabrication into targets.

The plutonium-238 oxide process was constructed over Section 6 of the canyon. The process is an improved version of the original plutonium-238 oxide line. The plutonium nitrate solution is converted to plutonium oxide powder by the oxalate precipitation followed by calcination. The cabinets associated with this line were equipped with significant external neutron and gamma shielding to protect the operating staff.

The scrap recovery module consists of two parallel lines of gloveboxes located over Sections 2 and 3 of the canyon. They provide the capability to introduce different types of scrap materials, sorting, dissolution, filtration, and transferring solutions to the canyon for recovery of the actinides and the associated support capabilities. The cabinet lines are equipped with water jackets and lead shielding over the gloves and lead glass on the windows. This equipment is provided to shield the operating staff from the gamma radiation.

Fabrication of Neptunium Targets in 235-F Building

Neptunium oxide from HB Line finishing operations was blended in Building 235-F with aluminum powder and fabricated into targets for irradiation in SRP reactors. Target fabrication was initiated in 1961 and consisted of a compacted blend of oxide and aluminum clad in an aluminum can. "Green" compacts, 3 inches long and 0.86 inches in diameter, were formed by pressing the blended powder in a tool-steel die at 19.8 tons per square inch (tsi) at ambient temperatures. These compacts were 90 to 92% of theoretical density. A double-acting press was used since it transmits equal force to both ends of the compact to give more uniform compaction. Complete densification during green compact fabrication was undesirable since some travel of the compact surface relative to the can wall during hot pressing is needed to provide a fresh metallic surface for bonding. Two compacts were loaded into an impact-extruded aluminum can, which was then closed by an aluminum cap. The assembly was loaded into an Inconel-X die, which was placed in a vacuum furnace consisting of a floating Inconel-X back-up die inside a stainless steel sheath wrapped with resistance heaters. This configuration is shown in Figure 4. The slug was heated to 600 to 620°C under a vacuum of <1,000 microns of mercury and then pressed at about 19 tsi to form a metallic bond between the compacts and the can wall and about 99% of theoretical density. All of these operations were performed in glove boxes with contained atmosphere to protect the workers. These target slugs were irradiated between 1961 and 1965 to produce plutonium-238.

In later development, as more neptunium became available for irradiation, the design of the targets was changed from target slugs to tubular targets, which improved heat transfer

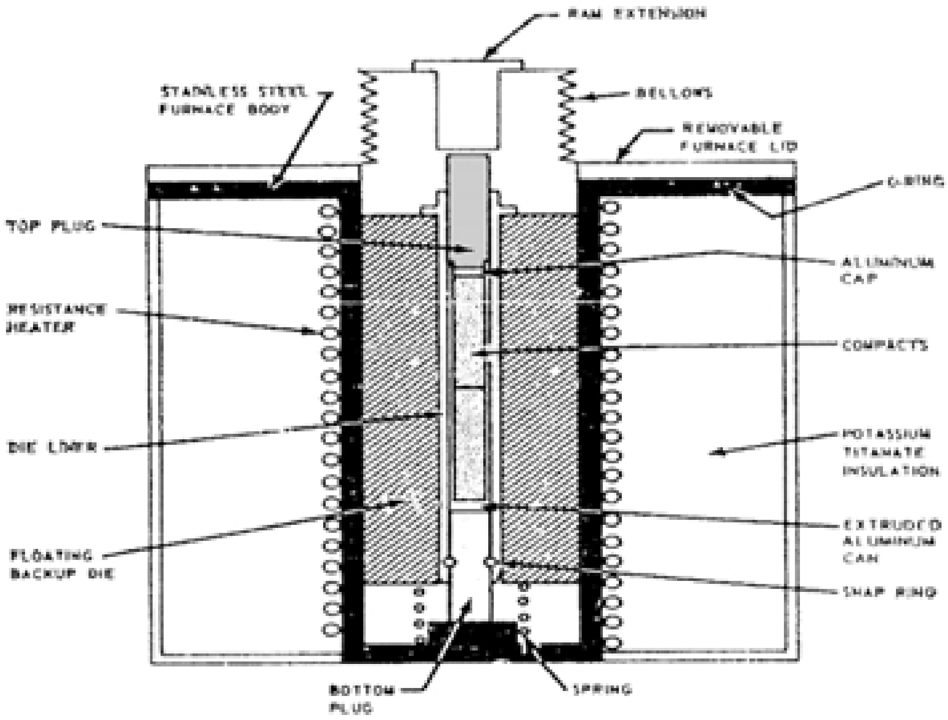

Figure 4. Hot-press-bond furnace for fabrication of Np target slugs

and increased plutonium-238 production and purity by allowing operation at higher neutron fluxes. Between 1966 and 1978, the design of these tubular elements changed three times. Billets containing the neptunium target were fabricated in Building 235-F, and the tubes were extruded in Building 321-M. Billets consisted of cylindrical inner and outer sleeves welded to the bottom fitting. Compacts were placed in the space between the two sleeves, and a top fitting with a breather tube was welded in place. This created a contamination shield, which allowed the billet to be removed from containment cabinets in Building 235-F and transported to Building 321-M for extrusion. The initial tubular targets were constructed with cylindrical compacts used in the target slugs, which on extrusion yielded tubes with ribbed target cores. Later compacts were redesigned into trapezoidal compacts to provide tubular targets with uniform neptunium core thickness. The different target design used differing amounts of neptunium so that the final tubular targets varied from 120 and 190 grams of neptunium per foot of active tube length, and the tube diameter varied between 3 and 3.7 inches. The initial cladding thickness was 0.065 inch and later was decreased to 0.040 inch as fabrication and irradiation experience showed these targets were safe for irradiation. Fabrication of neptunium target was continued until the SRS reactors were shut down in the early 1990s.

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Biographies

Harold J. Groh

Harold J. Groh has 47 years experience in nuclear programs. Employed at Savannah River Site from 1952 through April 1990 in professional and management positions, he has broad familiarity with DOE nuclear weapons complex and SRS capabilities, with particular technical expertise in nuclear fuel processing, waste management, and plutonium technology. He worked ten years as an independent consultant in numerous DOE programs including surplus weapons plutonium disposition, spent fuel management, chemical safety reviews, and high-level waste management. Mr. Groh has a Ph.D. in physical chemistry from the University of Rochester, and a B.S. in chemistry from St. Louis University.

W. Lee Poe, Jr.

W. Lee Poe, Jr. has more than 48 years of experience in providing technical and management support for large-scale nuclear projects including chemical processing, waste management, environmental protection. Employed by Du Pont Company and WSRC from 1951 through 1989, his experience includes operation of chemical reprocessing of irradiated spent nuclear fuels (F and H Canyon Buildings), purification and finishing of ^{239}Pu, ^{238}Pu, ^{237}Np, and U (depleted, natural, and enriched) (in FB and HB Lines and A Lines in both areas), target fabrication, heavy water production, and site and DOE complex planning. Process safety and operational assessments, environmental protection, safety and health protection, and a thorough understanding of the processes used in all areas of the SRS are an integral part of this experience. Specific responsibilities for ^{237}Np and ^{238}Pu processing were technical support within the various plant units for design, testing, start-up, and operation of all of the processes described in this paper. His experience also includes ten years consulting with DOE and private contractors associated with DOE nuclear programs in a wide range of areas. During this time, he has participated in public forums on waste management activities; risk management alternatives, comparisons, and ranking; environmental remediation; etc.

John A. Porter

John A. Porter has 42 years experience in research, development, and production in the defense nuclear industry. Employed by DuPont Company and Westinghouse Savannah River Company from May 1957 through July 1989 in professional and management positions at the Savannah River Site, his experience includes operation of chemical processing plants for irradiated fuels and targets (F and H Areas separations plants, ^{239}Pu metal production line, ^{238}Pu oxide production line, plutonium storage facility, plutonium recovery facilities, tritium facilities); operation of waste management facilities and analytical control laboratories supporting processing plants; manager of industrial hygiene and radiation protection functions; coordinator of environmental protection programs; and conduct/management of research and development activities relating to the above. With ten years independent consulting in DOE nuclear programs, his specialty areas include plutonium (and other actinides) processing, waste management, conduct of operations, personnel safety, and environmental protection. Mr. Porter has a Ph.D. in physical chemistry from Vanderbilt University and a BS in chemistry from Clemson University.

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Production of Pu-238 Oxide Fuel for Space Exploration

**D. Thomas Rankin, William R. Kanne, Jr., McIntyre R. Louthan, Jr.,
Dennis F. Bickford, and James W. Congdon**

Abstract

The Savannah River Site (SRS) made significant contributions to NASA space missions via the heat source programs that provide electrical power to satellites traveling outside of earth orbit. Electrical power was supplied by converting heat from the radioactive decay of plutonium-238. The Site produced the Pu-238 in its reactors, separated the plutonium in the canyon facilities, and, for the Multi-Hundred Watt and General Purpose Heat Source programs, formed plutonium oxide into pellets and encapsulated the pellets in the Building 235-F Plutonium Fuel Form Facility (PuFF). Technological innovations were behind the success of each of these steps in production. Using Pu-238 produced at SRS has allowed U.S. satellites to explore the solar system and beyond, sending remarkable pictures back to earth. Plutonium produced at SRS has powered 26 space missions. This report particularly addresses the technology for fabrication of plutonium oxide pellets and encapsulation of these pellets. Plutonium oxide pellets produced and encapsulated at SRS are now powering the Galileo satellite presently circling Jupiter and the Ulysses satellite presently in a polar orbit around the sun.

Introduction

Early in the U.S. space program, scientists recognized that an efficient source of power was needed for satellites. Batteries had the disadvantages of being very heavy and had lifetimes that were too short for deep space missions. Solar cells were in their infancy and could not operate at great distances from the sun. Technology to convert thermal energy into electricity was available, and the decay heat associated with radioactive materials could be the source of energy. Coupling radioactive decay heat with a thermoelectric converter became the power source of choice for satellites, particularly those that need to operate where solar energy is not plentiful.

Plutonium-238, produced and packaged at SRS, was selected as the heat source for thermoelectric power for satellites used for deep space missions. During the late 1950s, the Atomic Energy Commission requested that the Savannah River Site produce Pu-238 as a heat source for this application. To accomplish this new task, a massive interdisciplinary effort was required. Neptunium-237, the precursor isotope to produce Pu-238, was recovered from existing SRS processes where it had been produced as a byproduct in the reactor irradiation of uranium. The neptunium-237 was fabricated into targets and irradiated in SRS reactors to form Pu-238. The Pu-238 was separated from neptunium and fission products and processed to plutonium oxide (see “Development and Performance of Processes and Equipment to Recover Neptunium-244 and Californium-252” in this proceeding).

Plutonium-238 is currently supplying power for the Galileo spacecraft orbiting Jupiter and exploring this planet and its four moons; the Cassini spacecraft, which is on its way to a similar mission near Saturn; and the Ulysses spacecraft, which is in polar orbit around the sun. Long-term, deep-space missions, such as

Galileo, Ulysses, and Cassini, require a nuclear source to provide the electrical power needed to operate the instruments on board the spacecraft. Similarly, smaller radioisotopic heater units provide localized heat for electronic packages on the spacecraft.

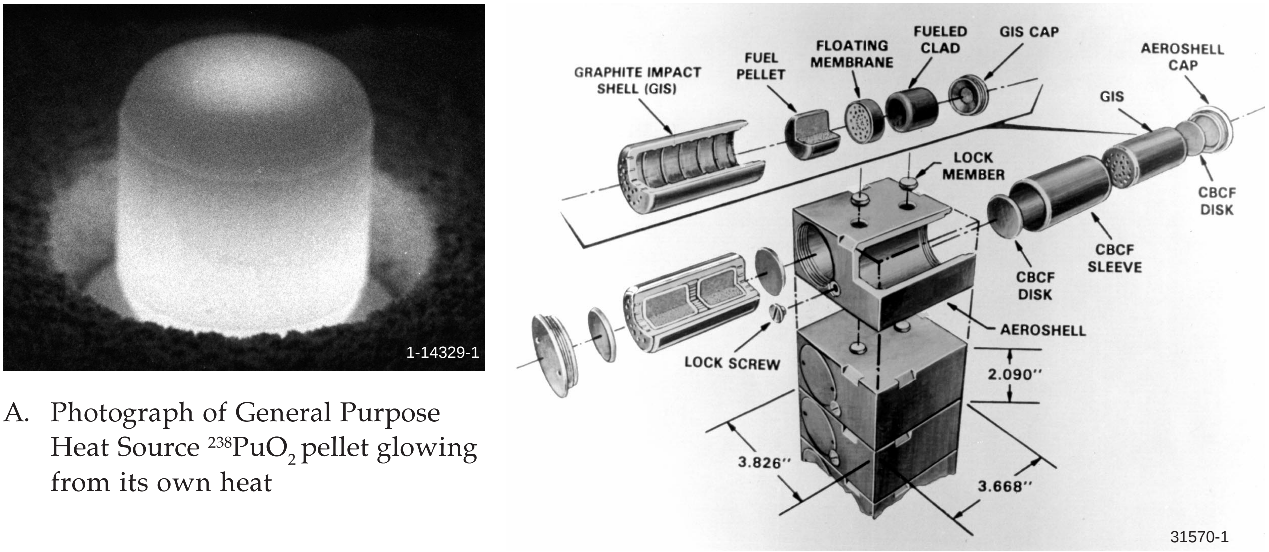
The Unites States has used, and continues to successfully use, radioisotopic thermoelectric generators (RTGs) to supply electrical power for deep-space missions. The RTGs consist of a nuclear heat source and a converter to transform the heat energy from radioactive decay into electrical power. Plutonium-238 serves well as the heat source because of its high power density (0.5 thermal watts per gram from alpha radiation), and its half-life of 87.4 years provides reliable and relatively uniform power over the lifetime of most NASA missions. The RTG is also a very reliable power source because it has no moving parts. All onboard electrical power to operate the cameras, collect the data, and relay information to Earth originates from the Pu-238 fuel produced at Savannah River.

Significance for SRS

Major scientific endeavors in space receive great benefit from technology at SRS. This technology produces and recovers Pu-238, processing the isotope into oxide, forming the oxide into pellets, and encapsulating pellets for long-term, high-temperature applications in U.S. space programs. Plutonium-238 produced and recovered at SRS supplied electrical power for all of the NASA deep-space probes, as well as the highly successful Apollo manned missions to the moon, the Galileo mission to Jupiter, the Ulysses mission around the sun, and most recently the Cassini mission to Saturn. In addition, data from instruments left on the Moon by the Apollo astronauts was transmitted to Earth for many years using electrical energy generated from the heat sources. The attention-getting Voyager 2 mission, which resulted in major discoveries about Jupiter, Saturn, Uranus, Neptune and their respective moons, was able to transmit, as a direct result of technology developed at SRS, information even after a 22-year, 15-billion-mile space odyssey.

The Galileo and Ulysses missions are particularly significant to SRS because these spacecraft use Pu-238 that was processed into pellets and encapsulated at SRS. The General Purpose Heat Source, containing 72 SRS encapsulated pellets, supplied the electrical power for these missions, which are ongoing at this time. The Galileo satellite (see Figure 1) has completed its initial objectives of exploring Jupiter and its four major moons. Additionally, it took the first photographs of the Earth and Moon together ever taken by an unmanned spacecraft, provided the first close-up photographs of an asteroid, and photographed the collision of comet Shoemaker-Levy 9 with Jupiter. The Galileo mission was extended for further studies of Jupiter's moon Europa. The Ulysses mission to fly over the poles of the sun was completed and provided a greater understanding of the behavior of sunspots, solar flares, solar x-rays, and solar radio noise. The Ulysses mission was also extended to investigate the high latitude properties of the solar wind. These extensions demonstrate the reliability of the Savannah River-produced Pu-238 pellets to provide power for extended space missions.

The Savannah River Site has played a major role in the U.S. space program. The successful use of Pu-238 in space allowed ventures outside of earth orbit that could not have been done without this reliable, long-lasting power source. The successful space missions that have occurred in the past and continue today were possible because of developments at SRS, beginning early in the history of the Site. Technologies used to develop Pu-238 for space missions include reactor physics, chemical separations, ceramic engineering, joining technology, materials engineering, and mechanical and electrical design. The extensive knowledge of the planets, the sun, and the moon that are available today are, in part, possible because



A. Photograph of General Purpose Heat Source $^{238}PuO_2$ pellet glowing from its own heat

B. Assembly of SRS $^{238}PuO_2$ pellets into General Purpose Heat Source module

C. Galileo satellite orbiting Jupiter (artist conception). GPHS Radioisotopic Thermoelectric Generators (on lower and upper booms) each contain 72 encapsulated $^{238}PuO_2$ pellets produced at the Savannah River Site using pellet fabrication and encapsulation processes developed at Savannah River.

Figure 1. Galileo mission to Jupiter powered by the General Purpose Heat Source, 1 of 26 space missions using SRS plutonium to provide power

of the ingenuity and devotion to scientific technology shown by many at SRS over the past 50 years.

Processing Pu-238 into Pellets

The preferred chemical form for a plutonium heat source is $^{238}PuO_2$, a ceramic (Rankin 1982). This face-centered cubic oxide offers excellent chemical stability, a high melting point (>2450°C), and chemical compatibility with its container material, an iridium alloy. The absence of phase changes in this material facilitates fabrication and enhances the integrity of the heat source during processing and use at an operating temperature of about 1350°C. A fabrication process, developed at the Savannah River Site, produced 150 g cylindrical pellets, approximately 2.7 cm by 2.7 cm, each of which generated 62.5 watts (thermal) at the time of production. Seventy-two of these pellets are contained in a single RTG that produces 285 watts of electrical power. The RTGs are compact (0.42 m in diameter by 1.13 m in length) and low in weight (55.5 kg), two requirements for efficient space systems. Two RTGs supply all the electrical power in the Galileo spacecraft.

The fabrication process for the cylindrical pellets consists of hot pressing a blended mixture of sintered $^{238}PuO_2$ granules prepared from calcined plutonium oxalate powder. Hot pressing provided the dimensional control needed and produced nominal pellet density of 84.5% theoretical. Both the granule sintering and hot processing conditions required close control to minimize cracking and to eject the pellet from the hot press die. A uniform pellet density and the distribution of large intergranuar porosity in the microstructure provide for the dimensional stability and the release of the decay helium (from alpha particles) required at the elevated use temperature.

A process suitable for pellet production was developed at SRS. This development was initially based on experiments at the Los Alamos National Laboratory and earlier pellet production at the Mound Facility. The SRS pellet production process, starting with calcined plutonium oxalate powder, consisted of the following steps:

1. Oxygen exchange to reduce neutron radiation by exchanging the naturally occurring ^{17}O and ^{18}O with ^{16}O by heating 5 hours at 800°C in ^{16}O.
2. Outgas for 1 hour at 1000°C in ^{16}O to free stored decay helium from the powder before particle sizing steps are initiated.
3. Ball mill for 12 hours at 100 rpm to produce a more uniform shape and an average particle size of 1.4 mm.
4. Granule formation by compacting the milled powder at ambient temperature to about 50% of theoretical density, sizing to <125 mm by forcing (with a roller) the material through a sieve.
5. Sinter 40% of the material at 1600°C and 60% at 1100°C for 6 hours in argon to densify the granules.
6. Sieve the densified granules to eliminate agglomerates that may form due to the self-heating of ^{238}Pu.
7. Blend the sintered and sieved material.
8. Vacuum hot-press at 1525°C and 19.4 MPa in inductively heated graphite dies to form the pellets.
9. Heat treat for 6 hours at 1525°C in an oxidizing atmosphere to assure dimensional stability, reoxidize to a O/Pu ratio of 2.00, and remove volatile impurities.
10. Vacuum outgas at 1500°C for 1 hour to reduce detrimental impurities and reduce the O/Pu ratio to 1.99 and thereby reduce material transport within the final capsule.

These ten process steps produced a chemically and dimensionally stable pellet that had the desired density, O/Pu ratio, porosity distribution, and other quality measures.

Encapsulation of Fuel Forms

Primary containment for each radioactive pellet is provided by a shell of iridium alloy (Kanne 1983). Iridium is a platinum-group metal that is compatible with plutonium oxide and has good strength and impact resistance at the 1310°C heat source operating temperature. Iridium is unusual in its inertness, high density, and rapid work hardening.

The containment capsules were thin-wall cylindrical shells that were 0.025-inch thick. A vent assembly in the end of the capsules allows the escape of helium gas from radioactive decay, but it does not allow escape of PuO_2 fines that may develop during the service life of the pellet. The welded butt joint was backed by a 0.005-inch thick foil of iridium to minimize the effect of weld heat on the PuO_2 pellet. A unique shipping container was designed at SRS for shipping the welded capsules to the Mound Facility for assembly into an RTG. The capsules were held in graphite felt within the shipping container. At the Mound Facility, the capsules were loaded into a graphite matrix before final assembly into the heat source modules.

A gas tungsten arc welding process was used to weld the girth of the iridium alloy capsules. The welding process was adapted from the Multi-Hundred Watt heat source welding process initially developed at the Mound Facility and briefly used at SRS prior to the General Purpose Heat Source program. The welding process produced a full penetration weld by welding at a relatively high speed of 30 ipm. Magnetic arc oscillation was used to promote the desired grain structure within the weld.

Welding the heat source capsules required a unique welding station for operation in the hot cell lines in Building 235-F. The welding equipment was computer controlled at a time when desktop computers were in their infancy. The welding station consisted of a turntable, an upper dead-weight-loaded positioner, and a horizontally mounted torch. Stepping motors actuated the three components. Welding current was supplied by a power source with a dual schedule programmer to accommodate tack welds and full welds. The computer automatically rotated the capsule through a series of three short tack welds and then the full closure weld. The flexibility of the computer-controlled welding equipment was a significant aid in developing the welding process and in creating an efficient production operation.

The welding operation was carried out remotely using manipulator arms and glove ports in the heavily shielded cells in Building 235-F. Remote operation was required because of the toxic nature of the plutionium-238 alpha-emitting radioisotope that was encapsulated. Welding speed was a compromise between higher speeds that produced centerline grain boundaries and slower speeds that resulted in through-wall grain boundaries. Welding current was a compromise between high current that caused the formation of a large columnar grain structure and low currents that could lead to lack of penetration. Small grain size with irregular boundaries was desirable. A hot cracking problem resulting from liquation of grain boundaries was minimized by four-pole arc oscillation. Hot cracking was eliminated in production capsules by introducing a state-of-the-art ultrasonic test that was used to cull out capsules with cracks and assured that only high quality welds were placed in service.

Plutonium Fuel Form Facility (PuFF)

The Pu Fuel Forms Facility (PuFF) was constructed in Building 235-F, adjacent to the Neptunium Billet Line. The Billet Line was the source of the Np-Al billets extruded for Mark IV production of Pu-238. The first floor of the PuFF consists of two remote manipulator lines (see Figure 2). The East Line conducts the Pu-238 powder receiving, processing, hot pressing and furnaces, and the West Line contains the welding of iridium cladding, decontamination, and welding of shipping containers. Five gloveboxes are attached to the maintenance side

Figure 2. Pluntonium Fuel Form Facility Hot Cell, Building 235-F

of the East Line, and the vacuum hot press. Also on the first floor is the Pu Experimental Facility, with full-size powder processing capabilities similar to the Puff East Line. An innovative inert gas system provided ventilation for the East Cell Line, as well as emergency and maintenance ventilation. A complete metallography glovebox line on the second level was used to examine pellet production and iridium sample welds. Final facility checkout was completed in 1977, and immediate production began on Multi-Hundred Watt spherical fuel. The facility was then reconfigured for a longer campaign of General Purpose Heat Source pellet production. Technical difficulties were encountered and overcome: rapid failure of glovebox gloves, highly mobile Pu powder, rapid pellet cracking, and underbead cracking of the iridium welds. The PuFF produced General Purpose Heat Source product that was acceptable when tested by high-speed impact at Los Alamos National Laboratory. The PuFF Facility was shut down at the end of the Ulysses fuel production, and the remaining NASA requirements have been met by Los Alamos.

Acknowledgments

This paper is dedicated to D. Thomas Rankin, friend, colleague, valued scientist, and one of the technical leaders throughout the SRS Pu-238 heat source program. Tom died October 23, 1999.

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Biographies

D. Thomas Rankin

Thomas Rankin was an advisory scientist and a program manager of the SRTC plutonium immobilization program at the time of his death in October 1999. Early in his career, he worked on the use of various isotopes for heat sources applications before playing a major role in the application of Pu-238 for the General Purpose Heat Source program. He received his B.S. in 1963 and his Ph.D. in 1967 in ceramics from Rutgers University. He was with SRTC since his graduation, except for duty as a military commander during 1968-70 at the U.S. Army Materials & Mechanics Research Center. He was a fellow and member of the Board of Trustees of the American Ceramics Society.

William R. Kanne, Jr.

William Kanne is a senior fellow engineer at the Savannah River Technology Center. He worked on encapsulation of MHW and GPHS heat sources during the late 1970s and 1980s. He has 30 publications and one patent on topics that include welding of irradiated materials, corrosion, and resistance welding in addition to welding of heat source capsules. Mr. Kanne is a graduate of the Johns Hopkins University with a B.A. in physics. He received a Ph.D. in metallurgical engineering from the University of Wisconsin in 1968, and he has been at Savannah River Site since that time. He is currently on the Board of Directors of the International Metallographic Society and is active with the American Welding Society, ASM International, TMS, and ANS.

McIntyre R. Louthan, Jr.

McIntyre Louthan is a senior advisory engineer in the Materials Technology Section of Savannah River Technology Center. He is the author of approximately 200 technical publications, editor of nine books and a fellow in ASM International. Mr. Louthan developed the lecture “Why Stuff Falls Apart”, which has been given over 200 times to colleges, universities, and civic and professional organizations. He has served as president of the International Metallographic Society, a member of the Board of Trustees of the National Youth Science Foundation, and chairman or co-chairman of 12 international conferences. He was a key reader for Metallurgical Transactions, a member of the editorial advisory board for Materials Characterization, and the series editor of Microstructural Science. Mac has given invited presentations throughout the U.S., Canada, Europe, and Asia and is a member of Sigma Xi, Alpha Sigma Mu, and Tau Beta Pi.

Dennis F. Bickford

Dennis Bickford is an advisory engineer at the Savannah River Technology Center where he is principally concerned with melter design and glass melting for DWPF and Idaho National Environmental Engineering Lab. Denny was Separations Technology manager for Building 235-F Operations during the startup of the Plutonium Fuel Form Facility and during production of the fuel for the Galileo and Ulysses missions. He received a B.S.M.E and B.S. in Metallurgy and Materials Science from the Massachusetts Institute of Technology and a M.S. in Metallurgy and Materials Science from Carnegie-Mellon University. Postgraduate studies include Ceramics Engineering at Clemson University. He is a member of Pi Tau Sigma mechanical engineering honorary society and Phi Kappa Phi. He is a fellow of the American Ceramic Society and president of the Board of the National Science Foundation Center for Glass Research.

James W. Congdon

James Congdon is a senior fellow scientist at the Savannah River Technology Center. He has worked at SRTC since 1977 in material and process development. He assisted in the development of the fabrication process for the General Purpose Heat Source and later developed several process improvements. Mr. Congdon is currently leading the SRTC effort to develop a process to immobilize excess plutonium. He received his B.S. in 1973 and his Ph.D. in 1978, both from Alfred University in Ceramic Science. He is a member of the American Ceramic Society.

Hydrides for Processing and Storing Tritium

Theodore Motyka

Abstract

The Savannah River Site (SRS) has 50 years of experience in handling and processing tritium for defense and other special applications. During the past 20 years, a new technology, metal hydride technology, was introduced to the tritium facilities. This technology dramatically changed the way tritium and the other hydrogen isotopes were handled and processed at SRS. Metal hydrides allowed tritium to be stored much more compactly and at much lower pressures, thereby minimizing accidental release and enhancing operational safety. The use of metal hydrides also simplified many of the processes, resulting in smaller and more efficient operations, which led to significant cost savings.

Multimillion-dollar cost savings have been realized in the existing tritium facilities at SRS by using metal hydride technology. Similar cost savings are expected in several of the new tritium projects. New tritium applications continue to be developed at SRS to ensure the reliability of our nation's tritium reserves and to support our nation's commitment to a strong defense.

In recent years, the Department of Energy and SRS have supported the development of "dual-use" metal hydride technology, which provides benefits not only for defense but also for future energy applications. SRS has collaborated on international energy programs aimed at demonstrating nuclear fusion as a potential, clean, and plentiful source of future energy. SRS has also partnered with government, industrial, and academic institutions to apply its expertise on metal hydrides to clean, non-polluting, hydrogen-powered energy systems. Benefits from these dual-use activities have allowed SRS to maintain its expertise in metal hydrides and have led to substantial cost savings for SRS facilities.

Introduction

What are metal hydrides?

While almost all metals can be made to react with hydrogen under some conditions, only a few metals do so "reversibly" at room temperature and near atmospheric pressures. These materials are generally referred to as "reversible" metal hydrides. Reversible metal hydrides have the ability to absorb and release hydrogen like a solid sponge (Sandrock and Huston 1981). They can do this over and over again. An analogy is that of a household sponge, which can absorb and release water as needed. Reversible metal hydrides can be either pure metal such as palladium, titanium, or zirconium. They can also be intermetallic compounds or alloys made up of two or more metals such as iron-titanium or lanthanum-nickel.

Reversible metal hydrides offer a number of advantages in storing hydrogen versus compressed gas or a cryogenic liquid. Hydrides have an extremely high volumetric density for hydrogen. That means that a lot of hydrogen can be packed in a very small compact space. In fact, most metal hydrides can store hydrogen several times more compactly than high-pressure gas and often more compactly even than liquid hydrogen. This is because the hydrogen atoms in a hydride are bound to metal atoms more closely than they can bind to themselves either as a gas or a liquid. Hydrides can store hydrogen at very low pressure, which affords a higher level of safety. Hydrides often react only with hydrogen, which makes them ideal for use in many separation processes. The disadvantage of hydrides is their relatively high cost and weight.

History of Metal Hydrides

While the ability of some pure metals to absorb hydrogen has been well known for over a 100 years, the discovery of a new class of intermetallic alloys that reversibly absorb and release hydrogen did not come about until the late 1960s. One of the first intermetallic alloys to be “hydrided” was iron-titanium. Brookhaven National Laboratory reported this in 1969 (Sandrock and Huston 1981). Iron-titanium was one of the first practical metal hydrides. It readily absorbs hydrogen at room temperature and is relatively inexpensive. However, iron-titanium also has some disadvantages. It can be easily poisoned by small amounts of oxygen, and substantial heating of the material is required the first time that it is exposed to hydrogen. The initial conditions, required of a metal hydride, to first absorb hydrogen are normally referred to as its “activation” conditions.

Around the same time that iron-titanium was being explored as a practical metal hydride, another important hydride material, lanthanum-nickel, was discovered. The hydride properties of this material were discovered entirely by accident (Sandrock and Huston 1981). The researchers at the time were working on developing permanent magnets when they stumbled on the hydrogen properties of lanthanum-nickel. This became a new and exciting reversible hydride material. Lanthanum-nickel has a high hydrogen capacity and readily absorbs hydrogen at ambient pressures. Furthermore it can be easily activated at room temperature without additional heating and is considerably more resistant to oxygen and other hydride poisons. In the early 1970s, researchers all around the word began exploring the properties of this new reversible hydride along with its many variations. It was discovered that substituting other metals for some of the nickel in the formulation could significantly change the hydrogen properties of the material. Scientists and engineers could now customize their own hydride materials and come up with operating conditions to match the needs of their application. No longer did they have to settle for a hydride whose properties were just in the “neighborhood”. An engineer could now select the right metal hydride material to meet a specific hydrogen storage temperature and pressure.

Commercial Applications of Metal Hydrides

Metal hydrides have been used for many years as chemical additives or reducing agents in chemical processes. But it was not until the advent of modern room temperature in the early 1970s that reversible hydrides spawned a wide variety of useful applications (Lynch 1991). These applications range from hydrogen storage, separation, and refrigeration to metal hydride batteries used in many of our notebook computers and other electronic devices.

NASA developed some of the first applications of metal hydrides. The objective was to demonstrate long-term hydrogen storage for potential use in space propulsion. Automotive hydrogen storage applications soon followed in several locations around the world. Metal hydrides provided a compact and safe method for storing hydrogen both for engines as well as fuel cell applications. In the 1970s, several metal hydride vehicles, including automobiles, vans, forklifts and even mining vehicles, were successfully demonstrated.

Other demonstrations involved using metal hydrides as compressors to deliver hydrogen at high pressures and as heat pumps and refrigeration systems. Hydride refrigerators usually employ pairs of hydrides with different operating pressures. Hydrogen is allowed to flow from one hydride to the other. The hydride, which is loosing hydrogen, naturally cools, and this cooling can be used to provide refrigeration. A waste heat source can be used to return the hydrogen to the first hydride allowing the cooling cycle to be repeated.

One of the most important recent applications of metal hydrides, Nickel Metal Hydride (NiMH) batteries occurred in the late 1980s. These batteries are now widely used in small electronic devices and are being developed for larger applications such as electric vehicles. Some of the benefits of NiMH batteries are that they are rechargeable, have a high-energy density, have no memory effect, and have low toxicity compared to traditional Nickel Cadmium (NiCd) batteries (Lynch 1991). The next generation electric automobile, the General Motors EV1™, is expected to operate on NiMH batteries.

Another important use of metal hydrides is in gas separation. Several companies have developed hydrogen purifiers using metal hydrides. The ability of hydrides to absorb only hydrogen in many gas streams makes them useful in many chemical separation processes. One of the areas where this has been used most successfully is in tritium processing applications.

History of Metal Hydrides at SRS

In the early 1980s, the Savannah River Laboratory began a major program to develop and utilize metal hydrides in its tritium production facilities. Metal hydrides turned out to be ideally suited for tritium handling and processing. Tritium is the radioactive form, or isotope, of hydrogen, produced for defense programs by SRS for over 40 years. Tritium behaves chemically very much like normal hydrogen, except that it is radioactive and decays over time to a form of helium. It should be noted that metal hydrides were used to process tritium at other Department of Energy sites prior to the 1980s. These typically involved using mostly uranium for tritium storage. This material has many drawbacks compared to the materials developed by SRS. Uranium hydride materials require high temperatures to remove the tritium, which leads to tritium permeating or escaping through walls of the container. In addition the pyrophoric or flammable nature of uranium led to many safety concerns with regards to its use at SRS (Ortman et al. 1985).

The 1980s

The first metal hydride small-scale test at SRS began in 1981. A few years later, the first metal hydride applications in the tritium facilities were introduced in a new tritium loading operation. The facility required both near-term storage and compression of tritium. The hydride material chosen for this application was lanthanum-nickel-aluminum (LANA). By substituting a small amount of aluminum for some of the nickel in the lanthanum-nickel hydride, it was found that the operating pressure of the hydride system could be controlled. Therefore, a higher aluminum content alloy with 6% aluminum by weight could be used for low pressure, safe storage of tritium, while a lower aluminum content alloy with only 2% aluminum by weight could be used to pump or compress tritium to higher pressures (Ortman et al. 1990). Another advantage of using the LANA metal hydrides for this application was that pure tritium was always delivered to the loading process (Nobile 1991). In traditional tritium pumping and loading applications, if the tritium stays in the system for a prolonged period of time, some of the tritium is converted to 3-helium. Tritium naturally decays to 3-helium, a non-radioactive form of helium gas at a rate of 5.5% per year. Though this process of radioactive decay is small, over an 8-hour shift, it can produce enough helium to effect the purity of the tritium. The LANA, metal hydride is able to retain all of the decay helium and release virtually pure tritium on demand.

Later in the mid 1980s, another metal hydride material was added to the SRS loading system to create a higher pressure, or second stage, compressor. The entire system was very simple and reliable with no moving parts other than valves.

Another early application of metal hydrides in the tritium facilities was as a pump/separator. In 1987, a metal hydride pump/separator was put in service to provide pumping to the cryogenic distillation columns, as well as to purify inert gasses (i.e., helium, argon, nitrogen) from the hydrogen isotope stream. Prior to the use of the pump/separator, the feed system to the cryogenic distillation columns, which are used to separate the different isotopes of hydrogen, always operated above ambient pressure. This was done to ensure that no gases such as nitrogen and oxygen could leak into the system. Since the cryogenic distillation columns operate at very low temperatures, any gases leaking into the system, other than hydrogen, could freeze out in the line and plug up the columns. The pump/separator used a very low-pressure hydride (palladium) that was able to efficiently separate the hydrogen isotopes from the other gasses and deliver the pure hydrogen isotopes directly to the cryogenic distillation columns. The pump/separator was also able to do this safely from tanks that were below atmospheric pressure, thereby improving the overall performance of the system (Nobile 1991).

Following the successful adaptation of metal hydrides in the existing facilities, a new tritium facility, based on more extensive use of metal hydrides, was planned. The new facility, originally called the Replacement Tritium Facility (RTF), began construction in 1986. The RTF was designed to take advantage of the latest technology to enhance operational safety, increase safeguards and security, and to minimize tritium releases to the environment. The facility was located underground to help prevent unauthorized entry. Thick, reinforced concrete outer walls combined with redundant safety systems provided protection against natural disasters (i.e. tornadoes and earthquakes) and assured that the facility could be safely shut down with no threat to the environment. Other technological improvements included using nitrogen gloveboxes to provide secondary containment for the tritium processes, and "dry" (oil and mercury free) pumps to eliminate generating a major mixed waste stream. The RTF also introduced laser cutting to replace a mechanical shearing systems for unloading tritium reservoirs and a state-of-the-art computer-based control system to improve product quality and process operations (Motyka 1992).

Of all the improvements in the RTF, the most significant new technology introduced was metal hydride technology. The use of metal hydrides to store isotopes in place of tanks substantially reduced the size of the overall facility, thereby significantly lowering the cost of the project. The size reduction also facilitated confining the process equipment into gloveboxes, thereby minimizing atmospheric releases. Other hydride applications for pumping, separating, purifying, and compressing hydrogen isotopes not only simplified process operations, but also improved the reliability of many of the plant operations.

The 1990s

The RTF became fully operational in 1994. To date all of the metal hydride applications introduced into the facility have met or exceeded all expectations. Metal hydrides were integrated throughout the entire gas handling process. In the RTF, metal hydrides are used to separate, store, compress, and purify hydrogen isotopes. A variety of pure metal and metal alloys were selected to meet the facility operating requirements. Each of these materials and applications was tested and evaluated by the Savannah River Technology Center (SRTC) prior to introduction in the RTF. Samples of metal hydride material were put into a long-term test program to ensure that the materials performed as expected over time. Both small- and large-scale performance tests on each of the metal hydride applications in the RTF were evaluated as part of the development program. Also a large pilot-scale test program, which integrated many of the individual hydride applications to determine how they would work together, was undertaken. The facility used for these integrated tests was also used to help instruct and train operators and engineers prior to RTF startup. The development and testing of the metal hydride systems by SRTC personnel played a major role in the successful startup and operation of the RTF.

Following the RTF, new tritium programs resulted in the additional demand for metal hydride technology. In the mid 1990s, the tritium facilities needed a new method for safely and compactly storing tritium for prolonged periods of time, 7-10 years. A new Hydride Storage Vessel (HSV) was developed to address this problem. The HSV used a form of titanium as the hydride material. Using this material allowed the excess tritium to be stored at very low pressure, which eliminated the need for an external cooling system to remove any of the tritium's heat of decay. The use of metal hydrides in this application provided a safe and compact solution to the long-term tritium storage problem and avoided the use of more dangerous and more expensive high-pressure gaseous storage.

Also in the mid 1990s, the tritium facilities became involved in several new projects. One of these was the Tritium Facility Modernization and Consolidation Project. The goal of this project, which is referred to as Tritium Consolidation, was to reduce the overall physical size of the tritium facilities and to upgrade and modernize its capabilities. Again, to achieve the project goals, new metal hydride applications were required. A new storage bed design was developed that eliminated the need for a forced heating and cooling system, thereby minimizing the size of the facility. Another process improvement was the introduction of another class of metal hydrides, often referred to as "getters". These materials have been developed to remove small amounts of impurities from gas streams. One of the major commercial uses is to provide extremely clean gas to the semiconductor industry. These getters are typically comprised of zirconium-based metal alloys. In Tritium Consolidation, these getters will be used to remove small amounts of water and hydrogen from nitrogen and other process flush gases. Finally, improvements to the metal hydride, isotope separation system, which separates tritium from the other hydrogen species, were also made. These improvements not only reduced the overall space required but also were estimated to save the project $20 million.

A new Tritium Extraction Facility (TEF) is being constructed at SRS to process the tritium-bearing rods produced in a commercial-type nuclear reactor. This newly produced tritium will be stored in metal hydride beds similar to those developed for the Tritium Consolidation Project. The TEF will also use a new hydrogen separator, which will greatly reduce the amount of gas that will need to be processed in the facility. The metal-hydride-based separator will separate the tritium and other hydrogen isotopes from the byproduct gases, which is mostly helium. The purified hydrogen isotopes will be measured in accountability tanks and then sent on to isotope separation operations. The waste gas will be further purified to remove any residual tritium contamination and then released. The hydride used in this process is palladium, which is also used in many of the RTF separation processes. The design of the separator, however, has been substantially improved by eliminating a large auxiliary heating and cooling system, thus making the unit much more compact and efficient.

Benefits of Metal Hydrides for Tritium Applications

Many of the benefits of using metal hydrides for commercial hydrogen as well as tritium applications have been described above. These benefits include safe and compact storage as well as efficient hydrogen separation and purification. While many of these benefits have commercial importance, they are perhaps more important in tritium applications. The use of metal hydrides in the tritium facilities has led to a major reduction in the size of the process equipment, allowing the equipment to be easily contained and isolated in gloveboxes. Virtually all potential tritium leaks can be captured, and the release of tritium to the environment can be avoided. Another advantage of metal hydrides is enhanced safety. Hydrogen and tritium can be stored on metal hydrides at low pressure.

This provides an added measure of safety when dealing with tritium, which is not only extremely flammable, but is also radioactive. Simplicity is another advantage of hydride technology. Hydrogen isotopes can be transferred from one location to another, or even compressed, simply by heating and cooling the metal hydride materials. This permits simple and reliable designs for tritium equipment, often with no moving parts other than valves. Since most valves can be operated remotely, many of the hydride systems can be easily automated and controlled. This is a very important advantage in a nuclear material handling facility.

Future of Metal Hydrides at SRS

As long as there is need for a strong nuclear deterrent, SRS will continue to play a major role in providing the tritium needed to support the nuclear stockpile. To support this mission, SRS will continually need to modernize and update its tritium handling facilities. A major part of the future modernization will involve metal hydride technology. In recent years, however, many of the metal hydride scientists at SRS have begun to look elsewhere for metal hydride development opportunities. The down sizing of the nuclear stockpile, coupled with the completion of several major tritium initiatives, has allowed SRS scientists the opportunity to see if the metal hydride experience gained from defense work could be applied to many of today's environmental and energy problems.

Fusion

One area that can benefit substantially from the transfer of metal hydride technology is the major worldwide effort to develop nuclear fusion as a future energy source. The United States, Europe, Japan, and Russia have collaborated on fusion energy research and development programs. One major program, the International Thermonuclear Experimental Reactor (ITER), has a goal of providing a large-scale demonstration of a fusion power plant. The preferred fuel for a modern fusion power plant is a combination of the hydrogen isotopes, deuterium and tritium (D-T). The product of a D-T fusion reaction is a tremendous amount of energy. Basically, the reaction is similar to what takes place naturally to power the stars and our own sun. The major byproduct is nonradioactive helium. The fusion reaction is not self-sustaining, like a fission reaction, and no long-lived fission products are produced.

For a fusion reactor to proceed, a substantial amount of tritium and deuterium will be required. Initially this tritium will be stored and eventually separated and recycled back into the reactor. Many of the processes being considered for these tritium operations are based on metal hydride technology. Metal hydrides offer the same safety and reliability benefits to the fusion program as they do for defense applications. Fusion scientists from around the world have developed several new metal hydride materials and applications. Japanese scientists have developed a new low-pressure, metal hydride material (zirconium cobalt) that has similar storage and operating properties to that of uranium metal. One of the major benefits of this new material over uranium is that it is less pyrophoric and much safer to use. Other metal hydride applications have also been proposed and evaluated. SRS and the other DOE organizations have actively supported fusion energy initiatives over the past 30 years. The fusion community can benefit immensely from long-term SRS experience with metal hydride and other tritium handling technologies. Most of the SRS value to the fusion program comes from its many years of safely handling tritium, which in a large part is due to its extensive use and reliance on metal hydrides. SRS can also directly benefit from support of fusion programs. Future fusion facilities will be using the latest state-of-the-art tritium handling equipment. Extensive research and development on new and better ways to handle and process tritium will be required for fusion facilities to operate safety and efficiently. Therefore SRS is in good position not only to contribute it expertise but also to learn from these future tritium facilities.

Hydrogen Energy

While the world waits for the development of future energy sources like fusion, a more near-term energy solution will be required. The U.S. dependence on foreign oil has increased substantially since the oil shortages of the early 1970s. The U.S. now imports more than half of its annual demand for oil. The energy problem is compounded even further on the world scene, where increasing development in third world countries will place an even greater demand on limited world energy supplies. How long will the world energy reserves last when the billions in Asia and other parts of the world demand the same automobiles, air conditioners, and home appliances found in the industrialized nations? Further complicating this issue is the fact that the majority of the world's oil reserves are located in politically unstable regions.

The debate on how long our energy supplies might last will continue but another perhaps even more important issue remains—*pollution*. Most of the major cities in the U.S. have serious and even life threatening air pollution and smog problems. When combined with the right atmospheric conditions, smog can seriously effect the health of the elderly and the very young in our cities. The situation around the world is much worse. Many major metropolitan areas in other countries are now restricting traffic and industrial operations during peak times of the day in an attempt to reduce the level of air pollutants. Another issue that also needs to be considered is global warming. While scientists continue to debate the extent of this problem, it cannot be disputed that the amount of carbon dioxide and other greenhouse gasses in the atmosphere has increased dramatically over the past century. This increase coincides with the increased use of fossil fuels. For these and many other reasons, alternative fuels need to become a larger part of the energy make up of this country and the rest of the world.

Many of today's potential energy alternatives—such as wind, solar, geothermal, hydro, and nuclear—produce only electricity. Storage of this electricity has been a problem. The development of advanced battery technology to store excess electricity has been disappointing. A better energy carrier is required to convert this electricity to something useful to meet our transportation needs. Hydrogen is the leading candidate. Hydrogen is the most abundant element in the universe, and, when used as a fuel with oxygen, it produces pure water as a byproduct with no pollutants. Very little pure hydrogen exists naturally on earth. Hydrogen on earth is found in the form of water or hydrocarbons such as oil, natural gas, and other organic materials. Most of the hydrogen produced today for industrial uses comes from processing natural gas and other hydrocarbons. In the future most hydrogen will come from electrolysis or photochemical reactions that convert water to its basic parts, hydrogen and oxygen. The hydrogen that is produced can then be used as a fuel in a direct combustion engine or in a battery-like device called a fuel cell.

One problem, however, still remains. How will we safely and efficiently store the hydrogen? One solution is metal hydrides. The same technology that SRS used to safely and efficiently store tritium and the other hydrogen isotopes for over 20 years can now be used to help the nation's and the world's energy problems. The ability of metal hydrides to store hydrogen compactly and at low pressures make them an ideal candidate for hydrogen storage systems on board future vehicles.

Dual-Use Hydrogen Technology

SRTC, funded by the Department of Energy's Office of Energy Efficiency and Renewable Energy (DOE-EERE), has recently developed several metal hydride-based systems for hydrogen vehicular applications. In April 1997, a hydrogen-powered bus called the H2Fuel Bus first rode down the streets of Augusta, Georgia. The H2Fuel Bus was a large-scale, demonstration project that enlisted the combined talents and efforts of commercial companies, academic institutions, and SRS engineers and scientists. The overall goal of the project was to develop a hydrogen-powered vehicle that employed SRS metal hydride technology. The metal hydride system onboard the H2Fuel Bus performed better than expected as it safely stored and delivered hydrogen to the bus's internal combustion engine during its testing and operational phases.

Another multi-partnered project, this time using a fuel-cell vehicle, was initiated in 1998. Fuel cells are a promising new technology that functions like a hydrogen-fueled battery. The advantage of a fuel cell is that it can generate power continuously as long as it is supplied with a source of hydrogen. Therefore, the fuel cell itself never looses its charge and does not need to be recharged. The goal of this project was to demonstrate and to eventually commercialize an industrial fuel-cell vehicle. The vehicle could be used in various indoor locations such as airports and warehouses to replace current electric-battery vehicles. The combined fuel cell-metal hydride vehicle can significantly outperform battery-powered vehicles and eliminate the harmful exhaust associated with gasoline-powered vehicles. A large part of the safety and reliability of the system is a result of using the SRTC-developed metal hydride system to store the onboard hydrogen.

Other projects currently underway at SRTC involve the use of fuel cell—metal hydride technology in mining vehicle applications. Increased regulation on the levels of carbon monoxide and diesel exhaust particulate in underground mines has forced the mining industry to pursue both battery and trailing cable electric alternatives. The performance of these alternatives, especially in very deep and long mines, has often been found to be unacceptable. A fuel cell–metal hydride alternative can be the solution by providing the range and power of a diesel vehicle without the harmful exhaust. Safe and low-pressure storage on solid metal hydrides will be the only acceptable hydrogen storage method for underground mines. SRTC is working with several commercial and academic partners in leading this effort toward a clean and efficient alternative vehicle for the mining industry.

The projects described above, along with other similar projects supported by SRS and DOE, have not only made an impact on our nation's energy future but have also helped to maintain the critical skills needed to support our nation's long-term defense mission. SRS scientists and engineers have been able to maintain their 50 years of expertise in the area of hydrogen/tritium storage and separation by continually taking on new challenges and opportunities. SRS is also better able to attract and retain the best scientific talent available by providing them with a stimulating and varied work environment. The benefits of supporting "dual-use", hydrogen/tritium technology at SRS cannot be overstated. Various improvements in recent tritium storage and separations systems at SRS came about as a direct result of technology developed for hydrogen energy applications. This has led to substantial cost savings and improved operations at the SRS Tritium Facilities.

Summary

The use of metal hydrides for storing and handling tritium and hydrogen isotopes has dramatically improved the overall safety and efficiency of the SRS tritium operations. Metal hydrides continue to be used in today's tritium facility to store, separate, purify, and compress hydrogen isotopes. The ability of metal hydrides to compactly store tritium at low pressure greatly reduces the potential for atmospheric releases. The simplicity of metal hydrides also improves the efficiency of many of today's tritium operations and helps lower operating costs. Metal hydride technology can offer similar benefits to future fusion and hydrogen energy systems. With the continued support of SRS and DOE, scientists at SRTC can develop new solutions to future hydrogen technology problems, thus, allowing SRS to maintain its critical tritium and hydrogen expertise well into the next millennium.

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Biography

Theodore Motyka received a BS degree from the Newark College of Engineering in 1973, a MS degree from Cleveland State University in 1975, and a Ph.D. from the University of Colorado in 1979 all in chemical engineering. In 1980 he joined the Savannah River Laboratory as a research engineer. Since 1989, Mr. Motyka has been a manager in the Hydrogen Technology Section of the Savannah River Technology Center supporting the development of tritium and hydrogen storage and separation processes. He and his group have numerous papers and hold several patents in this field. In the last two years, Mr. Motyka has been actively involved in the development and demonstration of hydrogen energy technology.

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Thermal Cycling Absorption Process—A New Way to Separate Hydrogen Isotopes

Myung W. Lee

Abstract

The thermal cycling absorption process (TCAP) is a semi-continuous chromatographic process for hydrogen isotope separation developed at the Savannah River Technology Center (SRTC) of the Savannah River Site (SRS) to support the tritium production and recycling operations for the national defense program. Hydrogen has three isotopes—protium, deuterium, and tritium. Among these, protium is known as normal hydrogen. Deuterium and tritium are heavier isotopes and are used in weapon systems. Deuterium and tritium fuse to form helium plus a free neutron, then releasing huge amounts of energy (1.7 trillion joules per mole). This is the source of energy for boost weapons. Fusion also serves another vital national interest—energy security. The problem is that the separation process required to supply tritium is costly, cumbersome, and inefficient. The TCAP method is compact, safer, and more efficient and an important technology for supporting the weapons program as well as for future fusion reactor fuel processing.

Introduction

Hydrogen has three isotopes—protium, deuterium, and tritium. Among these, deuterium and tritium are used in weapon systems, which requires the separation of deuterium and tritium in high purity. Isotopes have very similar chemical properties. Therefore, isotope separation is, in general, more difficult than chemical separation. Deuterium is relatively cheap because it occurs in nature in 150 parts per million. The rest is hydrogen. In nature, tritium is produced by cosmic bombardment and is only found in trace level. As a result, tritium is essentially a man-made isotope.

Tritium is radioactive and disintegrates to helium-3 and beta rays. A tritium nucleus has two neutrons and one proton while helium-3 has two protons and one neutron. During the radioactive decay of tritium, one neutron turns into a proton and beta rays. This decay half-life is 12.3 years (Souers 1985). (If the neutron were free, the half-life would be 12 minutes. The presence of two other nuclear particles stabilizes the neutron in tritium.) Because tritium reduces by half every 12.3 years, tritium can continually be produced and reprocessed to maintain defense capabilities.

Several technologies exist to separate gaseous hydrogen isotopes (Lee 1993). Cryogenic distillation uses liquid hydrogen (at −265°C). This operates at high density with high capacity. Thermal diffusion uses high temperature gas at 800°C. It is not an economical system for hydrogen, but a good system for purifying inert gas as such as helium, argon, or krypton. Both systems are large and occupy a three-story building. Another method uses the batch sorption columns based on gas chromatograph. This method has low capacity. A better method is desired, and TCAP meets that need.

Thermal Cycling Absorption Process Achievement

Development History

In 1981, TCAP-I was built at SRS. It consisted of a 5-ft column heated with electricity and cooled with cold glycol liquid, which was in turn cooled by dry ice. Tests proved the concept. A year later, TCAP-II, a 20-ft column, was built to generate scale-up parameters, with heating and cooling from a Freon™ compressor. The process control was further developed and plug flow reverser (PFR) was added to the system. PFR allows gas to flow out of the column without mixing and reverse the direction of gas flow back into the column again without mixing, maintaining the concentration profile. In 1982, a project was set up to replace the thermal diffusion column with TCAP (Lee 1982; 1983). To support the project, TCAP-III, a 40-ft column, was designed and tested at the Savannah River Technology Center using hydrogen and deuterium mixtures. Test results demonstrated the purity and capacity, which met the requirement of plant operation. In parallel to TCAP-III, an aluminum-block design was proposed by the engineering group. After a long study, the aluminum-block design was terminated because of fabrication difficulty.

In 1985, the Replacement Tritium Facility (RTF) (the current Building 233-H) was proposed. In this new facility, processes were all based on metal hydride. The thermal diffusion column (a three-story building) and storage tanks were replaced by TCAP and metal hydride storage beds, respectively (Heung 1985). SRTC built the Advanced Hydride Lab (AHL) to test new metal hydride technologies for the Replacement Tritium Facility, each unit operation, and interactions among units. TCAP-IV (stainless steel coil design) was installed in the AHL (Horen 1991). The heating and cooling system here was based on nitrogen gas. Since TCAP-III, no change has been made on the column length and the diameter that determines the product purity and the throughput capacity.

Thermal Cycling Absorption Process Principle

TCAP is a gas chromatograph in principle using palladium, but is operated in a semi-continuous manner. TCAP consists of a column packed with palladium coated on kieselguhr and a PFR packed with plain kieselguhr. Kieselguhr helps to reduce the pressure drop along the column and provides a large surface area of the palladium metal to get a fast isotope exchange reaction. One end of the column is connected to a PFR. A thin layer of palladium on kieselguhr readily absorbs hydrogen gas. It absorbs more hydrogen at lower temperatures then at higher temperatures. The equilibrium pressure at a given hydrogen concentration in palladium (in atomic ratio of hydrogen to metal, H/M) and at given temperature is known as "isotherm".

Palladium also has a very large isotope effect (Lee 1983; 1985; 1991) and preferentially absorbs the lighter isotope. This isotope effect is quantified by a separation factor, as defined by the ratio of the heavier to lighter isotopic concentration in the gas phase to the same ratio in the solid phase. In another words, it is a ratio of those two ratios. The separation factor is larger at lower temperatures. This separation factor is sometimes called the "single stage " separation factor. A column can have many stages. The overall enrichment factor of a given column is approximately the single-stage separation factor powered by the number of stages. Therefore, a larger separation factor and a longer column length give a better separation.

Thermal Cycling Absorption Process Operation

A mixed gas stream is fed at a fixed location in the middle of the column. The PFR is connected at one end of the column. An enriched stream is withdrawn at the opposite end of the column, and the depleted stream is withdrawn at the PFR end of the column. The column is thermally cycled by the heating and cooling system. During the heating in the regeneration cycle, the hydrogen gas is desorbed from palladium, and the pressure in the column increases. Then, the desorbed gas is transferred into the PFR. During the cooling separation cycle, the hydrogen gas is absorbed into palladium, and the column pressure decreases. At this point, the gas in the PFR is transferred back into column. As the gas flows through the column, the isotopic exchange occurs between the gas and the solid (Pd) phases. By the isotope effect described above, the heavier isotope is released from the solid and exchanged for the lighter isotope. The heavier isotope (tritium) migrates toward the far end of the column. The heavier isotope is enriched here and depleted at the PFR end of the column. During every cold cycle, separation takes place with the lighter isotope preferentially absorbing onto the palladium. During the desorption cycle, some of the separation gained is lost, and the column is regenerated. But since the separation factor is greater at colder temperatures, net separation is obtained after each complete absorption-desorption cycle. This net separation produces the product and raffinate purity.

In the total reflux mode, the gas is moving back and forth between column and PFR (no feed, no withdraw). After many cycles, a relatively sharp boundary is formed in the middle of the column. High purity isotopes are at the both ends. In the production mode of the operation, a small fraction of the mixture is fed. The product (heavy isotope) and raffinate are withdrawn. The fraction of the product stream to the total withdraw is determined by the concentration at the feed point.

TCAP has been operated in Tritium Operations. It achieved higher purity products. TCAP will replace cryogenic distillation in the near future. Los Alamos National Lab has planned to build TCAP for their tritium operations.

Design Consideration

The throughput capacity is proportional to the total column inventory of the hydrogen isotope and inversely proportional to the cycle time. The column inventory is directly proportional to the column length and to the cross-section area of the column. By increasing the column diameter by two, the system capacity increases four times.

The purity of the product and raffinate depends on the column length, the temperature difference between hot and cold, the throughput rate, and the feed concentration. Higher purity can be achieved by a longer column, smaller feed rate, or larger delta temperature. But the longer column has a higher pressure drop and longer time for gas transfer.

The separation factor of protium to tritium is much larger than that of deuterium to tritium. Shorter columns can achieve the same purity for tritium-protium separation as that of tritium-deuterium separation. With the same column length, the feed rate can be larger for tritium-protium separation than tritium-deuterium separation.

Process Models and Simulation

SRTC developed mathematical models and computer simulation programs for TCAP operation (Lee 1984). Quantitative relationships between column length, cold and hot temperatures, and feed rate to the product/raffinate purity could be calculated. This simulation package has been a valuable resource for design calculation and optimization of the operation. For example, the product and raffinate purities for D-T and H-T separations as a function of the feed rate could be calculated. Two operational temperature sets are in a nominal range (-40 to 150°C). The purity of product (tritium) and raffinate (deuterium) depends on the temperature set, feed rate, and the isotopic mixture. In addition, other variables are the feed location, the column length (number of stage), and the feed concentration. For engineering reasons, the high and low temperatures are limited. The capability of the simulation program serves well for project design.

Based on the given requirements such as throughput capacity and the purity of product and raffinate, the design parameters such as (1) column length, (2) column diameter, (3) feed location, and (4) hot and cold temperatures of heating/cooling system can be determined using the program. Some of the requirements can be achieved by adjusting the operational control parameters such as feed rate for a fixed TCAP system. The acceptable ranges of hardware and control parameters are essential pieces of information for the system design and can be obtained by the process simulation (Lee 1999).

TCAP Advantage

TCAP has a very small footprint. It fits into a glove box (the first barrier for radiation release), thus providing a safer method of handling radioactive materials. It is operated in a nominal temperature range. In the semi-continuous operational mode, TCAP can have a very large throughput rate. The process control is very simple. It is based on the mid-column concentration rather than desired product or raffinate purity. It has one feed stream and two output streams, one for product and one for raffinate. Therefore, TCAP requires no tank for intermediate cuts. Because of these advantages, TCAP is the choice of technology in the tritium process.

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Biography

Myung W. Lee received his Ph.D. in physical chemistry from Brown University. He has more than 30 years of experience in chemical dynamic, isotope separation, quantum chemistry, and material science areas. He published 30 technical journals and over 100 reports ranging of basic quantum mechanics to practical applications. He holds eight patents and over a dozen invention disclosures. His current interest is in areas of metal hydride and molecular modeling of materials.

Development of Resistance Welding Methods for Tritium Containment

William R. Kanne, Jr. and Robert J. Alexander

Abstract

A resistance welding process was developed early in the history of the Savannah River Site (SRS) for the critical application of sealing reservoirs filled with tritium gas. Resistance welding again became the welding method of choice for attaching new stems to reservoirs to allow their reuse after tritium decay necessitated removing the reservoir from service. The successful application of these two welding processes put SRS in the forefront of resistance welding technology. These successes, with no failures during service, provided the basis for future non-reservoir applications, the largest of which is the 5-inch diameter defense waste canister closure weld. The success of the pinch and reclamation welding processes also led to developing resistance welding to join the two halves of reservoirs, with potential to replace fusion welding processes altogether for reservoir fabrication.

Introduction

During the early months of 1956, a project was initiated at the request of the then Atomic Energy Commission to design and build a new tritium facility. This facility was to provide the necessary equipment to fill and seal capsules (reservoirs) designed by Los Alamos Scientific Laboratory (LASL) (as it was known in 1956). This project was the beginning of a unique engineering task to develop a technique that would seal tritium in capsules at high pressures.

Engineers at LASL and Du Pont Atomic Engineering Division (AED) in Wilmington, Delaware, investigated five methods for sealing the capsules:

- Ball check
- Projectile closure
- Conventional packed valve
- Ball expansion seal
- Pinched tube

The first two options were abandoned for either failure to obtain the required leak rate or for not producing consistent results. By the end of July 1956, development of the conventional packed valve closure, method 3, was abandoned because of more pressing work, the expense required for testing, and the cumbersome valve that would remain with the capsule.

The fourth method, the ball expansion seal, was developed during the investigation of the other methods. This method required a steel ball to be driven into a tapered plug tube, expanding it to seal the annulus through which the tritium passed during loading. In early July 1956, it was decided that the capsules for the startup would contain the ball expansion seal and initial capsules were fabricated by ACF Industries Atomic Energy Section in Albuquerque, New Mexico. However, this method was also abandoned because of time and cost restraints, leaving a pinched tube as the only viable option.

Pinch Welding Development

The challenge of loading and sealing reservoirs was therefore met by developing pinch welds to close the tube through which reservoirs are loaded. Prior to this time, there was no history of sealing tubing containing pressures much higher than 100 psi. The development criteria required sealing a tube containing gas at pressures orders of magnitude higher that previously accomplished .

The pinch weld is a type of resistance weld initially unique to SRS for high pressure closures. During pinch welding the tube is sealed by applying a force and an electrical current through electrodes that impinge on the tube. Initially these tubes were 1/80-inch OD by 1/16-inch ID and were made of 304 stainless steel. These dimensions are used today for most reservoirs. Tube material became 304L when the low carbon grade became available, and most reservoirs today are made from this material.

The early pinched tube welds were made by DuPont at the Mechanical Development Laboratory in Wilmington, Delaware, and at the TNX Facility at the Savannah River Plant. In October 1956, development of the pinch welding of tubes was assigned to the Weld Development Group of the Engineering Assistance Section in Building 723-A. Development of the pinch welding process was completed by mid 1957.

The initial welds were made using a manual (foot operated) spot welder. The electrodes used initially were made of copper with either spherical- or cylindrical-shaped tips. The production welding equipment was designed by Du Pont and fabricated at the Site. This equipment was unique in that the reservoir tubes were held between the horizontal floating electrode rams.

Pinch welding process refinement and improvement continued for many years. The process was adapted to reservoirs with tubes of different sizes, materials, and strengths. A capacitor discharge welder was used to seal tubes as small as 0.040-inch O.D. by 0.010-inch I.D. Tubes as large as 0.250-inch O.D. by 0.083-inch I.D. were welded on the production design pinch welders. Processes to achieve a hotter weld interface were developed, including the confined tube welding process that was initiated in production in 1974. This process minimizes the diameter of the pinched tube and produces melting in the center of the weld to minimize the effect of tube bore cleanliness.

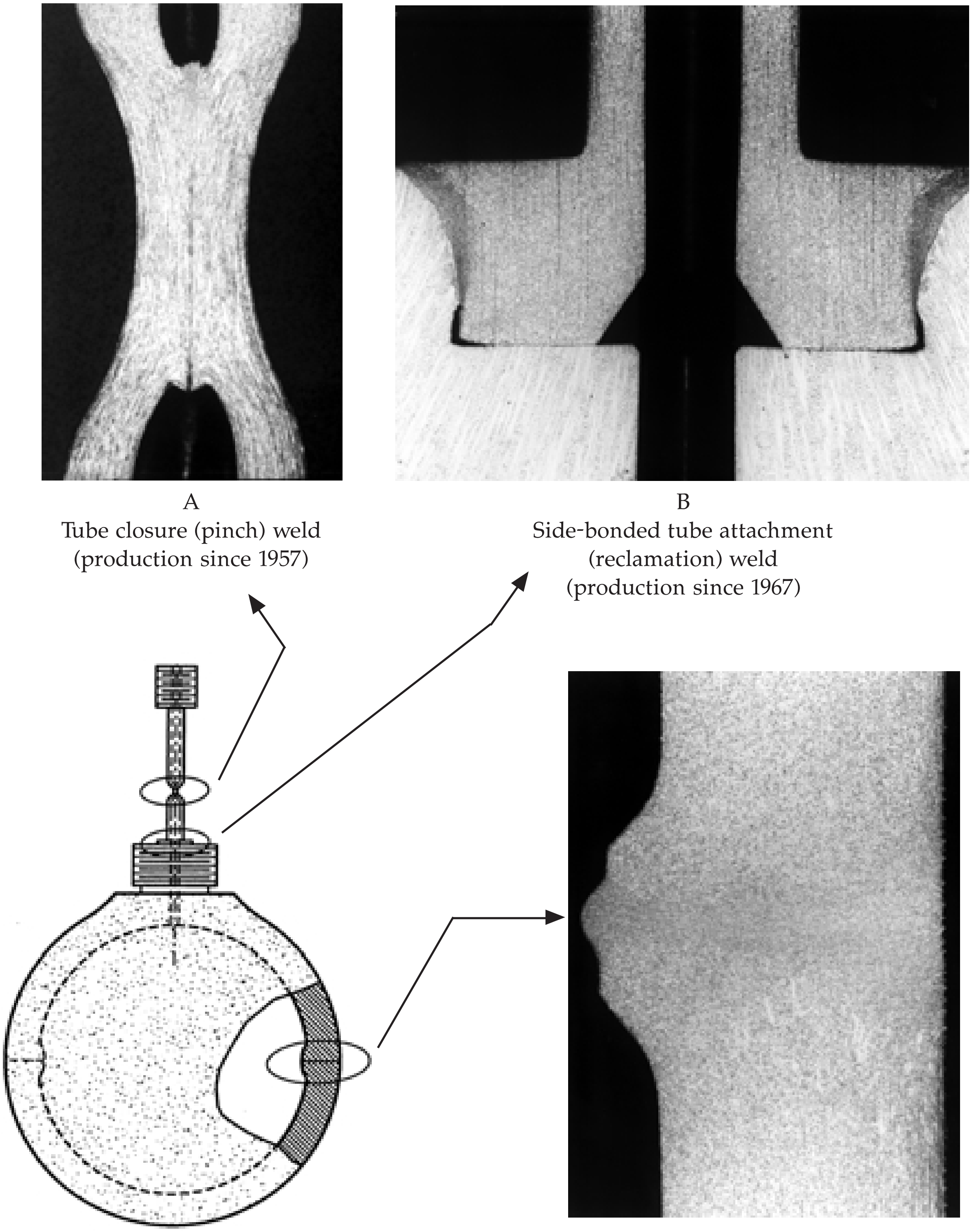
Developing the pinch welding solved a critical production problem at SRS and initiated a lasting technical expertise in resistance welding at the Site. Production pinch welding was carried out in loading lines in Building 234-H until 1994 when the loading mission was transferred to the new Tritium Facility in Building 233-H.

Reclamation Welding Development

The useful life of reservoirs is determined by tritium decay, which renders the weapon non-functional. Used reservoirs can be discarded and replaced. However, a large cost saving was realized by recycling, or reclaiming, reservoirs. Reclamation has been done in Building 238-H since its construction in 1969 by replacing the fill stem using a “reclamation” resistance weld. This method reuses reservoirs by filling the reservoir through a tube and pinch welding the tube.

The reservoir reclamation process was developed in the 1960s by Equipment Engineering in Building 723-A. Three processes were initially investigated during the developed program to reclaim reservoirs. Fusion (gas tungsten arc) welding, brazing, and resistance welding were all successfully developed for reclamation. These processes included both tube-to-tube welds and tube-to-base welds. The tube-to-base resistance welding process was chosen for production.

Reclamation by resistance welding was a novel process for attaching tubes to a part. The process requires machining and resistance welding capability in a glovebox facility. The old stem from the reservoir and a counterbore at the base of the stem location are machined and a new stem is resistance welded into the counterbore. The new stem has a “foot” that is oversized to the counterbore diameter by about 0.024 inch. During welding, force and electrical current are used to push the stem foot, which becomes hot from resistance heating, into the counterbored hole. A metallographic cross section of the reclamied stem is shown in Figure 1. Significant cost savings were realized by using the reclamation welding process.



A
Tube closure (pinch) weld
(production since 1957)

B
Side-bonded tube attachment
(reclamation) weld
(production since 1967)

C
Simulated reservoir

D
Girth weld
(currently in development)

Figure 1. Reservoir fabrication resistance welding processes (weld cross section)

In recent years, the manufacturing process for new reservoirs has incorporated the reclamation process to attach stems. The reclamation process is more cost effective, and stems are resistance welded to new reservoir bodies rather than machine stems as an integral part of the body. The past practice of drilling of the small hole through the stem when it is an integral part of the reservoir was very costly, and rejects were frequent. However, if stems are separate from the reservoir when they are machined, the most that will be lost from machining errors is the stem itself.

The production reclamation process has been very successful over the years with no failures during deployment. The first, and now preferred, reclamation process is called side-bonded because the weld forms along the sides of the stem foot. Other reclamation welds have been developed. For instance, a projection weld that requires no counterbore was developed with advantages in simplicity, minimal machining, and superior strength. Considerable tritium experience now exists for the projection tube attachment weld without a counterbore. This weld was applied in production using a shallow counterbore and was called a bottom-bonded reclamation weld.

A reclamation process combining a braze with a side-bonded resistance weld was developed and patented. The weld-braze technique created a weld that is much stronger than the side-bonded reclamation weld, but the technique was not applied to production since the projection weld has the same strength without the complication of a braze. An alternative reclamation process that uses a combination of laser drilling/welding with resistance welding was developed more recently and may find future application.

Reservoir Fabrication Development

Solid-state resistance welding is being developed to fabricate reservoir bodies. The production applications will be used at the DOE Allied Signal Federal Manufacturing & Technology plant in Kansas City, Missouri, where reservoirs are fabricated. The Kansas City plant purchased and installed large resistance welding equipment that will be used for reservoir fabrication. Initial development of girth welds to fabricate cylindrical and spherical shapes was carried out at SRS and continues today in support of applications at Kansas City.

Resistance welding has advantages of stronger welds, a simple process, fewer defects, and less sensitivity as compared to the fusion welding processes currently used to join the two halves of reservoir bodies. A simple butt weld joint around the circumference of the body components is all that is needed to form the weld. The girth weld is made by applying force on the weld joint and then passing a high electrical current through the weld joint. Fixturing is used to apply the force, to channel the current flow through the weld joint, and to align the two halves of the reservoir body during welding. Resistance to the electrical current at the weld joint, combined with resistance heating of nearby metal, increases the temperature at the weld joint to create a bond at the weld interface. No metal melting occurs. Welds up to 2.5 inches in diameter with a 0.3-inch wall have been produced. These welds are stronger and easier to make than the fusion welds they are designed to replace.

Beginning in 1984, vessels fabricated using solid-state resistance welding were placed in tritium storage in the Materials Test Facility, Building 232-H. The long-term compatibility of the fabrication welds for tritium service is being demonstrated with the storage tests. Vessels maintained their integrity during accelerated (71°C) storage for periods up to 12 years. Ten vessels were removed from storage and evaluated with no detrimental effects of the tritium observed. Over 170 vessel-years in storage were successfully completed.

Resistance Welding Fixtures, Controls, and Instrumentation

Mechanical and instrument development by the Equipment Engineering Department was an important part of the progress made on resistance welding. Successful application of unique welding processes required distinctive mechanical and instrument development.

Fixtures for pinch, reclamation, and girth welding all required adaptation to the unique configurations being welded. Copper alloy pinch welding electrodes were tipped with tungsten having a cylindrical radius that impinged on the tubes. Developing the confined tube pinch weld required fixtures with the added restraint on the sides of the tubes. Restraint was achieved using anvils made from Waspalloy in a fixture to align the tube with the electrodes and anvils. Reclamation fixtures required positioning the tube and reservoir to seat the tube in the counterbore while applying appropriate force and current. This was accomplished using a split electrode in an insulated sleeve for alignment. Girth welding uses large copper alloy electrodes in an insulating sleeve to align the two reservoir halves.

Advances in resistance welding controls and instrumentation were implemented throughout the history of resistance welding at the Savannah River Site. Data acquisition and weld control systems unique to SRS were developed, tested, and qualified in Building 723-A prior to installed in H-Area production lines. Emphasis over the years evolved from the early vacuum tube technology to the current computer-based data acquisition and control systems. During the 1990s, real-time digital sampling of resistance welding process electrical and physical parameters was introduced. This instrumentation provides accurate control and monitoring of process parameters during the welding operation. The technology was exported to Los Alamos National Laboratory to use with their resistance welding equipment. Additionally, feedback controls for welding current were tested, yielding a tenfold improvement in weld current variability. This progress has enabled production personnel to create more consistent and repeatable welds.

Historical Impact

The development of the pinch welding process in the early 1950s provided a robust method to seal tubes at high internal pressures. This technology was previously unavailable. Application of force with a shaped electrode and the passage of an electrical current through the tubes allowed sealing tubes with a range of sizes and materials. This closure method allowed the reservoir design laboratories great latitude in designing reservoirs. Pinch welding in its various forms has been used to produce reservoirs for 42 years without a field failure.

Both reclamation and pinch welding are unique applications of resistance welding. Developing tube attachment technology enhanced the Site expertise in resistance welding and provided a low-cost alternative to discarding used reservoirs. Technology and equipment for pinch and reclamation welding were transferred to other DOE sites over the years, in particular to Sandia National Laboratory, Los Alamos National Laboratory, the Rocky Flats Plant, and more recently, the Allied Signal facility at Kansas City.

Offshoots from the Site's resistance welding technology for tritium containment have been considerable. The largest spin-off is the plug weld used to close the canisters in the Defense Waste Processing Facility. The DWPF weld is 5 inches in diameter, the largest known application of resistance welding in the world. Additional SRS applications that relied on the resistance welding technology developed at the Site include the closure of Rabbit Capsules. These aluminum capsules were initially closed using a fusion welding process that exhibited many problems. These capsules, which contained iodine samples for irradiation in SRS reactors, were made routinely during the 1980s using the large resistance welder in Building 723-A. Another application was fabricating charging vessels that were used to expose mechanical test samples to tritium.

Work is ongoing in the area of resistance welding for containment of tritium. The development of large welds for fabrication of reservoir bodies continues. New equipment was recently installed at the DOE Kansas City Plant, the production site for reservoir fabrication, for applications using the large resistance welding processes developed at the Savannah River Site.

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Biographies

William R. Kanne, Jr.

William Kanne is a senior fellow engineer at the Savannah River Technology Center where his current work includes the use of resistance welding for tritium containment. He has 30 publications and one patent on topics that include welding of irradiated materials, welding heat source capsules, corrosion, and resistance welding. Mr. Kanne is a graduate of the Johns Hopkins University with a B.A. in physics. He received a Ph.D. in metallurgical engineering from the University of Wisconsin in 1968, and he has been at Savannah River since that time. He is currently on the Board of Directors of the International Metallographic Society, and is active with the American Welding Society, ASM International, TMS and ANS.

Robert J. Alexander

Robert Alexander is a principal engineer at the Savannah River Site in the Defense Program Tritium Engineering Division. His present work includes mentoring loading and welding system engineers and the development of new loading and welding processes for the Tritium Facilities. During his early years at Savannah River, he was a major part of the pinch weld development team. He later played a major role in the development of the reclamation welding process, and he holds a patent on resistance-braze welding. Mr. Alexander attended Florida State University and University of South Carolina at Aiken. He has worked at SRS for 46 years.

Evaporation and Storage of Liquid Radioactive Waste

Claude B. Goodlett

Abstract

The liquid wastes produced during the processing of radioactive materials at the Savannah River Site were initially stored in large underground tanks constructed of carbon steel. These liquid wastes were generated from the Purex process (for producing plutonium) and the HM process (for producing tritium). The liquid wastes were designated as high-level wastes and low-level wastes. As the production requirements increased with the resulting increase in liquid wastes, additional storage tanks and associated facilities were constructed. Since the waste volume was large and waste storage facilities construction was expensive, methods to reduce the cost of storing these wastes and increase the safety of storage were implemented. Storage tanks of differing designs and facilities to concentrate and to handle waste in the tank farms were constructed. These modifications resulted in significant cost savings and increases in liquid waste storage safety.

Liquid wastes produced during radioactive materials processing at the Savannah River Site were discharged to large underground tanks for interim storage. Processing these stored wastes is underway to convert them to a solid form for permanent disposal. A vitrified waste form will contain most of the radioactivity, and a concrete waste form (saltstone) will contain most of the chemicals. This paper addresses the interim storage of these liquid radioactive wastes from the initial processing of radioactive material, which began in 1954. During storage, these wastes were concentrated by evaporation to reduce their volume resulting in major economic savings and increase the safety of storage (Goodlett 1976a, 1976b).

Types of Waste

The liquid radioactive wastes from the separations plants (221-F and 221-H) are alkaline, with a dissolved solids content of 30-35 wt %. Two different processes were used to produce plutonium and tritium. Although there were variations in each process, blending these wastes in the large storage tanks and using similar chemicals in the two main processes resulted in using related processes to handle the waste.

Plutonium Production

Plutonium production used uranium metal in a rod or tubular form clad in aluminum. The aluminum cladding was dissolved from the uranium metal core in a solution of sodium hydroxide and sodium nitrate. The uranium metal core was then dissolved in nitric acid and processed through a solvent extraction/purification process (Purex process) to recover the plutonium and uranium. The wastes produced in the Purex process are of two general types; high-level waste, which contains sufficient radioactive fission products to produce decay heat at 0.5 to 5 Btu/(hr) (gal), and low-level waste, with fission product content 1/1000 to 1/100,000 that of the high-level waste, but still too high to discard to the environment. The low-level waste contains principally sodium aluminate and sodium nitrate from the caustic dissolution of the aluminum cladding on the irradiated fuel elements. The high-level waste, principally sodium nitrate with some sodium sulfate and sodium carbonate, contains nearly all of the radioactive fission products from processing the irradiated fuel elements. These liquid wastes are stored separately in underground storage tanks in the tank farms. During storage, both wastes separate into a layer of sludge and a layer of relatively clear supernatant liquid.

Tritium Production

Tritium production uses fuel tubes of enriched uranium-aluminum alloy with an aluminum cladding. After removal from the reactor, these spent fuel elements were processed to recover the enriched uranium. The entire fuel tube, uranium plus aluminum, was dissolved in nitric acid with mercury as a catalyst. The resulting solution was processed through a modified Purex solvent extraction process called the HM process. The waste produced in the HM process was similar to a mixture of the two types of waste from plutonium production.

Original Design Bases

The major liquid radioactive waste producers were the separations facilities located in the 200-F and 200-H Areas; specifically, the canyon Buildings 221-F and 221-H. Small quantities of liquid wastes from the Savannah River Laboratory and from the production reactors were sent to the 200-F Area.

200-F Area

The 200-F Area was originally provided with eight 750,000-gallon underground tanks constructed of carbon steel and designated as Type-I tanks. These tanks were of a cup-and-saucer design with the storage tank totally enclosed inside a 5-foot-high short steel tank, which served as a saucer. This entire unit was contained in a massive concrete tank. These tanks were coil cooled to remove heat from radioactive decay. The tank-in-tank arrangement would contain any radioactive material that might be spilled or leak from the primary tank. The concrete tank provided radiation shielding from the tank contents, was a vault to prevent water from contacting the carbon steel primary tank and saucer, and added seismic protection. An annular space between the concrete vault and primary tank was supplied with warm air to remove any moisture that might cause primary tank corrosion and facilitated visual inspection of the primary tank. As demonstrated during operation, the dry warm air in the annulus dried waste that seeped through small cracks in some of the primary tanks.

Liquid radioactive waste from Building 221-F flowed by gravity to a diversion box in the tank farm, where the waste was routed to one of the eight storage tanks. Since the waste contained only dissolved solids, the velocity of waste in the stainless steel waste transfer lines did not have to be controlled; however, the waste transfer lines were built without low points that would allow solids to settle and plug the transfer line. The Head End precipitation step in the separations process produced a solid manganese dioxide (MnO_2) cake. To prevent the transfer of solids to the tank farm, this cake was dissolved with gluconic acid. Adding of this organic acid was later discontinued when it was determined to be unnecessary. Radioactive waste from SRL and any other locations was trucked to an unloading station in 200-F before transfer to Building 221-F, where it was combined with waste from the 200-Area Laboratory (Building 772-F) prior to transfer to the tank farm.

200-H Area

Since the original concept was for Building 221-H to back up Building 221-F, only four 750,000-gallon underground tanks were provided. These tanks were identical to the Type-I tanks constructed in 200-F. To accommodate increased plutonium production, Building 221-H was placed in operation, and four additional liquid waste storage tanks were constructed. These carbon-steel storage tanks (Type II) were similar to the Type-I tanks, but held one million gallons. Because of water table considerations, this group of four tanks was constructed at a higher elevation than the original Type-I tanks. Consequently, the liquid waste had to be transferred by gravity feed and pumping.

The liquid radioactive waste flowed by gravity from Building 221-H to a diversion box in the tank farm, where it was routed to one of the four Type-I storage tanks. Since the elevation difference required that the waste had to be pumped to the four Type-II storage tanks, a second diversion box and three pump tanks were added to the original set of four Type-I tanks. All waste transfer lines and the pump tanks were constructed of stainless steel.

Second-Generation Waste Storage Tanks and Evaporators

To reduce the costs of storing these ever-increasing waste volumes, facilities were constructed in the tank farms to concentrate the stored waste and reduce its volume. The storage tank design was modified to reduce storage costs. Four uncooled waste tanks with a centrally located evaporator were constructed in the F-Area Tank Farm (Taber 1959). This facility became operational in 1960. These were the first facilities to concentrate the liquid radioactive waste in the waste tank farms. A similar facility containing four uncooled waste tanks with evaporator were constructed in the H-Tank Farm (Taber 1960); this facility became operational in 1963.

Tank Farm Evaporator

In the mid to late 1950s, laboratory work at Brookhaven National Laboratory with nonradioactive wastes from the Purex process showed that the low-level waste, principally a mixture of sodium aluminate, sodium hydroxide, and sodium nitrate, could be evaporated to reduce volume. Experimental work on the design of an evaporator suitable to evaporate this waste was done by the Griscom-Russell Company in conjunction with Brookhaven National Laboratory. This work showed that a chemical scale would form on the surfaces of the steam coils but could be removed by the design of a unique evaporator.

- It featured a steam chest with the heating tubes installed in a bent condition. This allowed the tubes to flex when supplied by alternating water and steam, thereby causing the scale to flake off of the heating surface.
- The lower portion of the evaporator was conically shaped to allow insoluble solids and the scale to settle.
- Since the contents of the evaporator were at the boiling point and design criteria did not allow bottom openings on vessels containing radioactive materials, a steam lift was used to remove the concentrated waste from the evaporator. A steam lift is a simple device consisting of an open pipe into which steam is injected at the lower end. The steam reduces the density of the concentrated waste in the pipe, allowing atmospheric pressure to carry the waste up the tube and out of the vessel (Goodlett 1963).

Uncooled Waste Tanks

The heat load in the low-level waste was sufficiently low that cooling coils would not be required in waste storage tanks that only contained low-level waste. Eliminating the cooling coils resulted in significant cost savings. Since these uncooled waste tanks would only be used to store low-level waste, the low radioactivity level in this waste did not require the tank-in-tank containment that was necessary for the high-level waste. These Type-IV storage tanks were 1.3-million-gallon underground tanks constructed of carbon steel. These single-wall carbon steel tanks were encased in a blown-on concrete shell. Prestressed steel reinforcing bands were used to support and prevent cracking of the blown-on concrete. This type construction eliminated the annulus between the steel tank and the concrete tank, which allowed for moisture removal and visual inspection of the primary tank.

Although this type of construction was justified when these tanks were built, later experience with cracking and waste leaking through the walls of some of the double containment tanks indicated that a single wall tank would not provide the protection to prevent waste loss to the environment that was present in the tank-in-tank steel tank. However, no single wall tanks have leaked waste to the environment. There was no annulus to allow inspection of the waste tank.

Waste Transfer Facilities

Installing evaporators in the tank farm was the first step in changing the waste storage areas into processing facilities. This change has continued to date. The evaporators are fed soluble liquid waste from one of the waste storage tanks using one of two systems: (1) a steam jet (this method of transferring waste was used in the separations buildings), or (2) a feed pump (an adaptation of a standard deep-well jet-pump system). Because the use of steam jets added water to the waste while the objective of evaporation was to remove water, later feed systems utilized a feed pump (Goodlett 1968d, 1972).

The concentrated waste from the evaporator is different from the waste discharged to the waste tank farm from the separations buildings in that some or all of the waste would solidify on cooling and could plug the transfer lines handling the concentrated waste. To prevent this pluggage, it was necessary to keep the transfer line from the evaporator to the receiving tank as short as possible, well-sloped, and insulated.

Third-Generation Waste Storage Tanks and Evaporators

As the production of nuclear materials continued, additional waste storage tanks and support facilities were needed. These needs resulted in the construction of additional waste storage tanks with a modified tank-in-tank design (Type III). The limitations of the single-wall tanks (Type IV) were recognized, and no more tanks of this design were constructed. Laboratory studies continued to improve the methodology to concentrate the high-level wastes from the Purex and HM processes to reduce volume and increase the safety of storage. Twenty-nine of these Type-III tanks (10 in 200-F Area and 19 in 200-H Area) and two additional bent-tube evaporators (one in 200-F Area and one in 200-H Area) were constructed. Also, facilities were installed to feed the waste to the evaporators and then transfer the waste from the evaporators to the waste tanks. In addition, a pipe line to transfer soluble waste between 200-F and 200-H Areas was installed, a distance of 2.5 miles (Goodlett 1968c).

Stress-Relieved Waste Tanks

Experience with small stress-corrosion cracks occurring in the primary waste tank in some of the Type-I and Type-II tanks resulted in the design of a new waste storage tank, Type III. These 1.3-million-gallon tanks were constructed of carbon steel and were similar to the tank-in-tank design of the Type I and II tanks except that the outer tank was a full-height tank providing two barriers of steel. They were annealed by heating the primary tank after construction to relieve any stresses present. Some tanks have insertable coils that were added through tank risers after construction. However, most of these tanks have installed cooling coils like the Type-I and -II tanks. These tanks had an annulus and a concrete outer tank. Since they were constructed over a number of years, the designs were modified as new knowledge was acquired.

Evaporation of High-Level Wastes

Experimental work carried out in the Savannah River Laboratory showed that all the wastes in the tank farms could be concentrated by evaporation to result in a solid when stored at room temperature (Goodlett 1968a). This process has been utilized in the tank farms.

The de-cladding waste from the Purex process is ideal for concentration. This aqueous waste contains no solid phase at its boiling point. A large difference exists between the boiling point of the solution and the temperature at which a solid phase is precipitated. When this solution is concentrated by a factor of 3.4, the hot concentrate is fluid, and a solid phase appears only when the solution is cooled 60ºC below its boiling point. The concentrated waste solidifies completely when cooled to room temperature.

Purex waste is more difficult to concentrate than decladding waste. When concentrated by more than a factor of 2.5, this solution contains a solid phase at the boiling point. The solid phase is initially a sandy, white precipitate of sodium carbonate and sulfate. Sodium nitrate is also crystallized as the solution is concentrated further or is cooled. When concentrated by more than a factor of about 3, the solution contains a large amount of solid phase at the boiling point. However, the concentrate does not solidify completely while cooled to 23ºC; about one-third remains liquid. Although further concentration can produce a material that will solidify completely on cooling, the amount of solids present near the boiling point is too high to be handled in the bent-tube evaporator without causing pluggage. Purex waste cannot be concentrated sufficiently by evaporation in one stage. However, this waste can be eventually reduced to a solid by four stages of evaporation, each stage followed by cooling and partial crystallization in a waste storage tank. The volume reduction obtained is 3.4.

HM waste behaves similarly to Purex waste with the exception that only three evaporation passes and successive coolings are required.

Transfer of Concentrated Wastes

Results from the SRL experimental data showed that the Purex and HM wastes could be evaporated sufficiently to generate a waste that solidified completely after cooling. However, this concentrated waste must be transferred over distances of hundreds of feet to waste tanks at elevations equal to or higher than the evaporator. All this must be done while maintaining a temperature close to its boiling point to prevent the settling of the undissolved solids present in the concentrated waste. Tests showed that simulated concentrated waste slurries containing at least 20 vol % solids could be pumped through a 2-inch pipeline if the bulk velocity of the waste was maintained at 1.6 ft/sec or higher (Goodlett 1968b). Based on these data, several transfer loops were installed to transfer aged alkaline waste (after concentration in a tank farm evaporator) to distant underground waste storage tanks. The circulation rate in these tank farm transfer loops is 3 to 5 ft/sec; two or three times the velocity corresponding to the onset of pluggage. These systems have operated successfully since initial startup in 1967.

Overall Effect of Waste Concentration in Waste Tank Farms

The evaporation of the liquid radioactive waste in the tank farms has significantly reduced the volume of stored waste. For example, in late 1986, the 77 million gallons of waste that was generated by operations in 200-F and 200-H Areas had been reduced to 32 million gallons, a reduction factor of 2.4 (Goodlett 1986). This volume reduction was less than achieved in the laboratory because all of the waste had not been concentrated to a solid. If this volume reduction had not occurred, additional waste storage tanks would have been required. Construction of these additional tanks would have increased the land area that contained radioactive facilities and would also have required costly subsequent decommissioning.

The waste transfer line between the 200-F and 200-H tank farms increased the safety and reduced the cost of waste storage by transferring waste between the areas.

Evaporation of the waste also increased the safety of waste storage because waste with a higher solids content was less prone to leak through any cracks that might develop in the steel wall of the storage tanks.

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Biography

Claude Goodlett graduated from Clemson University with a BS Degree in chemical engineering in 1954. He worked for Du Pont at the Savannah River Site from graduation until he retired in 1989. His areas of work were primarily in the processes and equipment for the processing of irradiated nuclear fuel and handling and storing radioactive wastes. He worked in both research and production functions.

He served as Chairman of the Savannah River Section of the American Institute of Chemical Engineers and Chairman of the Joint Council of the Engineering and Scientific Societies of the Central Savannah River Area. He has authored or co-authored 22 papers.

After retiring from Du Pont, Mr. Goodlett has worked as a consultant for Los Alamos National Laboratory, Brookhaven National Laboratory, Battelle Pacific Northwest Laboratory, Ebasco Constructors, Stone and Webster Engineering Company, and Westinghouse Hanford Company.

The Defense Waste Processing Facility, from Vision to Reality

Chris T. Randall, Lou M. Papouchado, and Sharon L. Marra

Abstract

When the Savannah River Plant began operation in the early 1950s producing nuclear materials for the national defense, liquid, highly radioactive waste was generated as a byproduct. Since that time, the waste has been stored in large, carbon steel tanks that are buried underground. In 1960, one of the tanks developed a leak, and before recovery measures could be taken, about 25 gallons of radioactive salt solution overflowed the secondary liner and seeped into the soil surrounding the tank. Significant improvements to the tanks were made, but constant surveillance was still required. Thus, the opinion began forming that storing the mobile, highly radioactive waste in tanks was not a responsible long-term practice. So, in the late 1960s, the Savannah River Laboratory began research to find a suitable long-term solution to the waste disposal problem. Several alternative wasteforms were evaluated, and in 1972, the first Savannah River waste was vitrified on a laboratory scale.

Introduction

When the Savannah River Plant (SRP) began operation in the early 1950s, highly radioactive waste generated during the production of nuclear materials for defense needs was stored in large, nominally 1-million-gallon, underground storage tanks. This mode of storage, which had been the practice at SRP's sister facility, Hanford, in Richland, Washington, has been judged to be a safe and effective way to isolate the hazardous radionuclides from the environment. However, in 1960, one of the tanks developed a leak, and, before recovery measures could be taken, about 25 gallons of radioactive salt solution had overflowed the secondary liner and seeped into the soil surrounding the tank. Although improvements to the waste storage tanks were made, including extending the height of the secondary liner to the top of the tank, constant surveillance was still required. Thus, the opinion began forming that storage of the mobile, highly radioactive liquid waste in tanks was not a responsible long-term practice.

In the late 1960s, the Savannah River Laboratory (SRL) began research to find a suitable solution to the waste disposal problem. Several alternative wasteforms were evaluated in terms of product quality and fabrication reliability. And, in 1972, SRL first vitrified actual Savannah River waste on a laboratory scale. Previously, Pacific Northwest Laboratory had studied waste vitrification since the early 1960s, and they provided substantial support during early development efforts at SRL.

By the mid-1970s, the Du Pont Company, then prime contractor at the Department of Energy's (DOE) Savannah River Plant, began to develop a vision of constructing America's first vitrification plant to immobilize the high-level radioactive waste stored in the SRP waste tank farms in borosilicate glass. This vision was later championed by Du Pont as a vitrification plant called the Defense Waste Processing Facility (DWPF). This plant was viewed with conviction as a timely step to close the nuclear fuel cycle and assist in developing a national nuclear waste disposal policy. Today, the DWPF processes Savannah River High Level Waste (HLW) sludge, turning it into a solid, durable wasteform of borosilicate glass. The DWPF is the world's largest vitrification facility, brought to reality through over 25 years of research and 13 years of careful construction, tests, and reviews at a cost of approximately $3 billion dollars.

The Vision

The vision embraced by Du Pont to immobilize SRP's highly radioactive liquid waste in borosilicate glass was an ambitious one. In the late 1970s, the Department of Energy recognized that there were significant safety and cost advantages associated with immobilizing the high-level waste in a stable solid form. However, at the time, there was not a consensus among leaders in the nuclear community regarding the wasteform or the process. Wasteforms had been studied since the 1960s, and, in the 1970s, a national and international consensus towards borosilicate glass was building. To reach a conclusion, the early studies were expanded and formalized to evaluate about 20 different wasteforms, including synthetic rock, tailored ceramics, glasses, and cement. This research confirmed that the radioactive species in the waste were bound chemically in the borosilicate glass matrix, making it a very durable wasteform. By 1979 borosilicate glass was clearly emerging as the top wasteform, based on an optimum combination of cost to produce, risk to people and the environment, and likely public acceptance it provided. France had selected borosilicate glass as the wasteform for a plant at Marcoule to immobilize high-level waste. England, Germany, and Japan were seriously considering this wasteform for immobilizing their high-level radioactive waste. Early in 1982, the choice of borosilicate glass for disposal of SR high-level waste (HLW) was endorsed by independent consultants engaged by Du Pont and the Department of Energy.

Although safety and protection of the environment were substantial drivers for the DWPF facility, the high cost of the storage tanks was an additional, and very tangible, incentive to construct the DWPF as soon as possible. After years of research, the wheels were set in motion in December 1981, when E. G. Jefferson, chairman of the Board of the Du Pont Company, in a letter to Edwin Meese, Counselor to the President, urged the Administration to support project funding. In accordance with the NEPA (National Environmental Policy Act) process, an Environmental Impact Statement was prepared for the facility, as well as an Environmental Assessment of the alternative wasteforms, and a Record of Decision (in December 1982) on the wasteform was issued. This Record of Decision was endorsed by the Environmental Protection Agency and several independent review groups. The Nuclear Regulatory Commission (NRC) also reviewed the document and offered no objection.

Early efforts by SRL to further develop the emerging technology focused on engineering calculations to define an integrated conceptual flowsheet, wasteform development to optimize processing and durability characteristics, and melter development. Experiments to define and demonstrate processes to prepare the waste for vitrification and to treat the offgas from the melter soon followed. The development and demonstration of the DWPF process was accomplished on a small scale in SRL's shielded cells, using actual waste and on a large scale using simulants in pilot facilities. Summarizing these developments, the first Technical Data Summary for the DWPF was issued in August 1978. The accompanying flowsheet was much different than the process we see operating today, demonstrating how the process has evolved.

Evolution of the DWPF Process

From the days of its inception until a few months before radioactive startup, the DWPF process evolved in response to the need for cost reductions, discoveries during development, and safety problems in the supernate pretreatment process. It is a tribute to the commitment and innovation of the entire team that supported and operated the DWPF that these changes, particularly those occurring after non-radioactive commissioning tests began, were accomplished with minimum impact to the vitrification mission.

The DWPF process was designed for Savannah River HLW, generated during processing fuels from the Site's nuclear reactors. Over 30 million gallons of the waste is stored in 51 carbon steel tanks in the form of settled sludge and saltcake. Neutralized to inhibit corrosion of the tanks, the waste contains a large fraction of non-radioactive chemicals and nearly every element in the periodic table. Because of this, the DWPF more resembles a complex chemical process than a nuclear process. To minimize costs, the overall strategy for vitrification of Savannah River HLW has always been to separate most of the non-radioactive salts from the radioactive constituents and dispose of this material in a less expensive manner than vitrification.

In the earliest flowsheets, the DWPF received a single, blended waste stream consisting of insoluble solids (sludge) and soluble salts (supernate). The sludge is the principal concern because it contains over 60% of the total radioactivity and essentially all of the long-lived radionuclides, which present over 90% of the hazard to man. The sludge was to be washed with water and centrifuged in the DWPF. The salts from the sludge were then to be combined with the supernate.

Although by weight the supernate is almost all non-radioactive salt, it contains most of the cesium in the HLW, and traces of strontium, which are highly radioactive. In the early flowsheets, these contaminants were to be removed by ion exchange and the decontaminated supernate was to be sent back to the Tank Farms to be evaporated to semi-dry saltcake and stored in decommissioned waste tanks. The radioactive stream from ion exchange was to be combined with the washed sludge and vitrified. The glass would be cast and sealed in stainless steel canisters and stored in a building onsite until a federal repository became available. This proposed process would immobilize the 30 million gallons of HLW in approximately 1 million gallons of glass, containing essentially all of the radioactivity, and several million gallons of decontaminated saltcake. Refinements to this early process were made, but the cost estimate, at $4 billion, was more than Congress would appropriate. The challenge was to cut the cost to less than $1 billion.

Serious efforts were therefore begun to reduce the cost and increase funding flexibility for the facility. Blending the sludge and salt streams in the Tank Farms was eliminated and two separate streams, salt and sludge, were sent to two separate DWPF facilities that could be funded in stages. An ion exchange facility decontaminated the salt stream, and a vitrification facility immobilized the radionuclides. Also, the cost of vitrification was significantly reduced by changing from two calciner-melter trains to one slurry-fed melter. But further cost reductions were needed, and research and design improvements provided opportunities.

The Melter

The melter, the heart of the vitrification process, was selected via evaluation of several different options. Considered the most unproven portion of the required technology, an emphasis was placed on melter development from the beginning. The first choice was a joule-heated ceramic melter. U.S. developers, Germany, and Japan favored this type melter over the inductively heated Inconel melter used by the French because of longer melter life and improved control of product composition. Du Pont had experience with high temperature, refractory lined reactors, making them cylindrical to increase reactor life. The first melter was therefore designed in a similar manner, and that design was not altered throughout development. This was a departure from the less expensive rectangular melters tested at other sites in the U.S. and Germany. The optimum materials of construction were identified early to be Monofrax K-3 refractory and Inconel 690 electrodes, heaters, and piping.

In August of 1980, SRL started up the first DWPF pilot melter. The design incorporated a spray calciner coupled atop the melter and top-entering electrodes to heat the glass via the joule effect. Horizontal resistance heaters in the vapor space initially heated a glass charge to the point that it would conduct joule current from the electrodes and to provide additional heat to boost the melt rate during production. Early testing, however, foreshadowed difficulty with remotely operating a spray calciner. This, and a substantial cost saving that could be realized by removing the calciner and decreasing the building height, prompted consideration of a change to an innovative process being studied by the Germans, feeding the waste and frit directly to the melter where evaporation, calcination and melting would take place. This was called the Slurry-Fed Melter.

At about the same time, an alternative process, the In-Can Melter, in which a waste/frit slurry was fed directly into a storage canister and melted by an induction heater was considered. Both the In-Can Melter and Slurry-Fed Melter were tested in pilot scale, and while the In-Can process had several advantages, including simplicity, it presented a problem in ensuring uniform glass quality. The slurry-fed melting process proved viable, however, and was therefore taken forward as the new design basis.

Several half-scale pilot melters were tested at TNX, SRL's semi-works facility, using simulated waste, as were several small-scale melters. A small joule-heated melter was also installed in SRL's shielded cells where actual waste samples were vitrified. Several unique design features were demonstrated through testing on these melters, including pouring by creating a vacuum in the cansiter and cooling the offgas via a device named an Offgas Film Cooler. The Offgas Film Cooler, invented by SRL and designed by the Du Pont Engineering Department, prevented pluggage of the offgas line exiting the melter, and is now used worldwide on slurry-fed melters. Pneumatic agitation to boost the melt rate was also tested in pilot melters. However, bubbler life was short due to corrosion and/or errosion by the hot glass, and agitation was abandoned.

Later, a one-tenth-scale pilot melter was built to test a more complete simulation of the waste, including mercury and noble metals. This melter, called the Integrated DWPF Melter System, or IDMS, was fitted with full feed preparation and offgas systems, which were constructed of design basis materials for corrosion evaluation. IDMS's primary mission, however, was to determine the fate of noble metals in the system, which tend to precipitate, creating a conductive path that would short the joule electrodes. In total, research melters at SRL, now called the Savannah River Technology Center (SRTC), poured over a million pounds of glass while refining and demonstrating the DWPF melter design and the DWPF process.

Supernate Decontamination

Removing the radionuclides from the salt component of the waste, however, proved to be a greater challenge than vitrification of the sludge. The original process employing ion exchange required a large canyon building (slightly larger than the vitrification plant). Commercially available elutable ion exchange resins were first proposed. With these, the cesium in the eluate had to, in turn, be loaded onto a zeolite for combination with the sludge due to the large eluate volume and lack of compatibility with the vitrification process. Because the facility was first estimated at over $1 billion, all development efforts were focused on reducing the size and cost of the canyon. Several improved resins were evaluated, and new resins were developed. A resorcinol-formaldehyde resin with significantly improved properties was one of the new resins. This resin reduced the size of the columns and the canyon, but the process was still very expensive.

A breakthrough occurred when a precipitation process using sodium tetraphenyl borate to precipitate cesium was developed. Because this process had potential to be implemented in existing waste tanks, it could provide savings on the order of $800 million. The process, called In-Tank Precipitation, was successfully demonstrated in an actual waste tank in 1983 and adopted as the preferred process. This was the breakthrough needed; accumulating all the cost savings, the estimate for the vitrification plant came in at just under $1 billion, and the DWPF project was funded.

However, in 1995 when the In-Tank Precipitation facility was started up, a larger-than-expected benzene release was observed. Extensive studies showed that a temperature-sensitive catalytic decomposition of the main reactant, tetraphenylborate, was responsible for release of the flammable benzene. Because the In-Tank Precipitation facility could not cost effectively meet the safety and production requirements for the high-level waste system, Westinghouse Savannah River Company (WSRC) suspended operations, and a study was initiated to evaluate alternative processing options.

DWPF, however, was designed with sufficient flexibility that the vitrification process could operated on flowsheets processing sludge only, sludge and salt, or salt only. DWPF is currently running a "sludge-only" flowsheet, bypassing the cell that hydrolyzes the tetraphenylborate precipitate to remove benzene for more efficient melter operation.

A Quality Product

Meanwhile, the development of the borosilicate glass product continued, as did the development of the regulations governing its quality. The Nuclear Waste Policy Act of 1982 mandated that all high-level waste would be sent to a federal repository for disposal. In 1985, the president ratified a decision made by the Secretary of Energy to send defense high-level waste, including the canistered wasteforms (stainless steel canisters filled with borosilicate waste glass) from the DWPF, to a civilian repository. The Department of Energy, recognizing that start-up of the DWPF would considerably precede licensing of a repository, instituted a Waste Acceptance Process to ensure that these canistered wasteforms could be accepted for eventual disposal at a federal repository.

Representatives from the repository projects and the wasteform producers developed preliminary waste acceptance specifications that identified requirements for the canistered waste forms. These early specifications were initially developed by SRTC and HLWM management personnel in support of DOE. The specifications eventually evolved into the Waste Acceptance Product Specifications for Vitrified High-Level Wasteforms (WAPS).

The WAPS require the DWPF glass wasteform to be more durable than an environmental assesment glass as measured by the product consistency test (PCT). The PCT involves placing crushed glass in sealed vessels filled with water for 7 days at 90°C. The leachate is then analyzed for the elements B, Na, and Li to determine the glass durability. These elements have been shown to bound the leach rate of the radionuclides.

Since DWPF does not have the ability to recycle unacceptable glass, and it is impractical to hold up the process for a 7-day test, it is desirable to control the process prior to vitrification. The DWPF ensures an acceptable glass product by controlling the melter feed composition. A correlation between composition and PCT results has been developed for use in control of the vitrification process. In addition to controlling the glass durability, as measured by the PCT results, it is necessary to ensure that the glass viscosity and glass liquidus temperature are within acceptable ranges. Correlations between these glass properties and composition have also been developed. These glass property correlations are embedded in the Product Composition Control System (PCCS) along with statistical algorithms to appropriately account for measurement error. The PCCS is the tool used by DWPF engineers to judge acceptability of the melter feed in each batch. Feed batches will be transferred to the melter only after there is confidence that they will produce acceptable glass. Occasionally a glass pour stream sample is taken to confirm acceptability.

Construction

Construction of the DWPF was no small feat of accomplishment. For protection of workers from radiation and contamination hazards associated with the HLW, and for protection for the public in the event of accident or natural disaster, the DWPF process is contained within a reinforced concrete building with 3-foot-thick walls. Processing cells within the building also have 3-foot-thick walls, and operations are conducted remotely. A robust ventilation system with redundant fans and emergency power ensures that air flows within the building are from clean, occupied areas into areas containing the process and are exhausted through a giant, underground sand filter outside the process building. Filters within the facility combined with the sand filter remove essentially all radioactivity in the ventilation air, even in the event of the worst imagined accident scenarios. Designed by Bechtel and constructed by Morrison Knudsen, the DWPF was built to withstand earthquakes and tornadoes with a functional lifetime well in excess of the 20 to 25 years required to immobilize all the HLW stored at the Savannah River Site. The facility contains 71,000 cubic yards of concrete and 10,500 tons of reinforcing steel. The 10-foot-thick concrete foundation mat is reinforced by 2-1/4-inch diameter reinforcing steel.

Groundbreaking for the DWPF occurred in 1983. Estimates in 1985 forecast project completion for September 1989, and radioactive startup in January 1990. However, as discussed later, radioactive startup was not to be until March 1996. Perhaps the greatest difference in the adjusted schedule and this early schedule lay in the time required for commissioning. The complexity of regulations and the degree of rigor required in commissioning combined with technical and engineering challenges to extended commissioning from a few months to a 5-year activity.

Startup Testing

Prior to the start of Radioactive Operations in 1996, DWPF underwent an extensive Startup Test Program. This test program consisted of Integrated Water Runs, Chemical Runs, Waste Qualification Runs, and Proficiency Runs. On a tight schedule, DWPF began functional checkout as sections of the plant were turned over to Operations. Integrated Water Runs, which tested piping and equipment up to the melter, were completed in 1992. During Cold Chemical Runs, simulated feeds and raw materials were introduced into the facility and the first batch of melter feed was produced. Melter heatup, initiation of melter feeding, and the first glass pour were completed in 1994.

The Waste Qualification Runs portion of the DWPF Startup Test Program was completed in 1995. During Waste Qualification Runs, varying feed compositions were used to demonstrate that the DWPF could control the glass product over the range of waste compositions expected. Simulated waste was transferred into the DWPF and processed using the same methods to be used for radioactive waste. Fifty-five canisters were produced during these tests, and the glass and canistered wasteforms produced were extensively characterized. The results of this characterization were the principal data that demonstrated the DWPF's ability to comply with the WAPS. In total, 80 canisters of simulated glass were produced during the Startup Test Program.

Following Waste Qualification Runs, Proficiency Runs were completed in which two batches of melter feed were produced performing all operations as though the feed was radioactive. The WSRC Operational Readiness Review (ORR) was completed, followed by the DOE ORR, and in March 1996 the DWPF was ready for Radioactive Operations. That month the first transfer of radioactive sludge arrived in the DWPF canyon building. The sludge was prepared according to well practiced procedure, and the glass frit was added. After concentration it was moved into the melter, and the first radioactive glass was poured into a canister. Canister decontamination and closure welding were completed, and the first canister of Savannah River HLW glass was moved in the Glass Waste Storage Building in May 1996.

A Worldwide Milestone, a Vision Realized

Early in the year 2000, DWPF is processing the second waste tank (macro-batch) of radioactive sludge. The first macro-batch yielded 495 canisters from 420,000 gallons of sludge. In January 2000, almost four years after beginning processing, the world's largest radioactive waste vitrification facility produced its 3 millionth pound of waste glass product, which is a new production milestone worldwide. Thus, the vision of building a plant to safely immobilize Savannah River high-level waste has clearly been realized. With the continued commitment of the Department of Energy and the Westinghouse Savannah River Company, the legacy of the remaining HLW at Savannah River will be processed into a stable borosilicate glass wasteform by 2028.

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Savannah River Site Waste Vitrification Projects Initiated Throughout the United States: Disposal and Recycle Options

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Abstract

A vitrification process was developed and successfully implemented by the U.S. Department of Energy's (DOE) Savannah River Site (SRS)[1] and at the West Valley Nuclear Services (WVNS) to convert high-level liquid nuclear wastes (HLLW) to a solid borosilicate glass for safe, long-term geologic disposal. Over the last decade, SRS has successfully completed two additional vitrification projects to safely dispose of mixed[2] low-level wastes (MLLW) (radioactive and hazardous) at SRS and the Oak Ridge Reservation (ORR). SRS, in conjunction with other laboratories, has also demonstrated that vitrification can be used to dispose of a wide variety of MLLW and low-level wastes (LLW) at SRS, ORR, Los Alamos National Laboratory (LANL), Rocky Flats (RF), Fernald Environmental Management Project (FEMP), and Hanford Waste Vitrification Project (HWVP). SRS, in conjunction with the Electric Power Research Institute and the National Atomic Energy Commission of Argentina (CNEA), have demonstrated that vitrification can also be used to safely dispose of ion-exchange (IEX) resins and sludges from commercial nuclear reactors. In addition, SRS has successfully demonstrated that numerous wastes declared hazardous by the U.S. Environmental Protection Agency (EPA) can be vitrified (e.g., mining industry wastes, contaminated harbor sludges, asbestos containing material [ACM], Pb-paint on army tanks and bridges). Once these EPA hazardous wastes are vitrified, the waste glass is rendered non-hazardous, allowing these materials to be recycled as glassphalt (glass impregnated asphalt for roads and runways), roofing shingles, glasscrete (glass used as aggregate in concrete), or other uses. Glass is also being used as a medium to transport SRS americium (Am) and curium (Cm) to the Oak Ridge Reservation (ORR) for recycle in the ORR medical source program and use in smoke detectors at an estimated value of $1.5 billion to the general public.

The Global Materials Cycle

Raw materials taken from the earth to produce a wide variety of products and processes must be disposed of safely back into the earth once declared as a waste (see Figure 1). The only other option is remediation for recycle into new products or new end uses. Technologies have been developed by the U.S. Department of Energy's (DOE) Westinghouse Savannah River Technology Center (SRTC) to convert many hazardous and/or radioactive wastes to a solid stabilized glass via the process of vitrification. The vitrification technology has been shown to render hazardous wastes to be non-hazardous, convert non-hazardous sludges, asbestos, etc., into recyclable products or reusable raw materials, or both.

If a waste cannot be recycled due to its radioactive content, then it must be safely disposed of back into the earth (see Figure 1). Stabilizing such wastes into glass by fusing the waste with glass-forming oxides (SiO_2, Na_2O, B_2O_3) at elevated temperatures in an electric melter[3] atomistically bonds the hazardous and/or radioactive species in the solid glassy matrix ensuring safe disposal for thousands (10^6) years. In addition, large volume reductions (up to 97%) allow for large associated cost savings for such wastes during interim storage, shipping, and long-term permanent disposal.

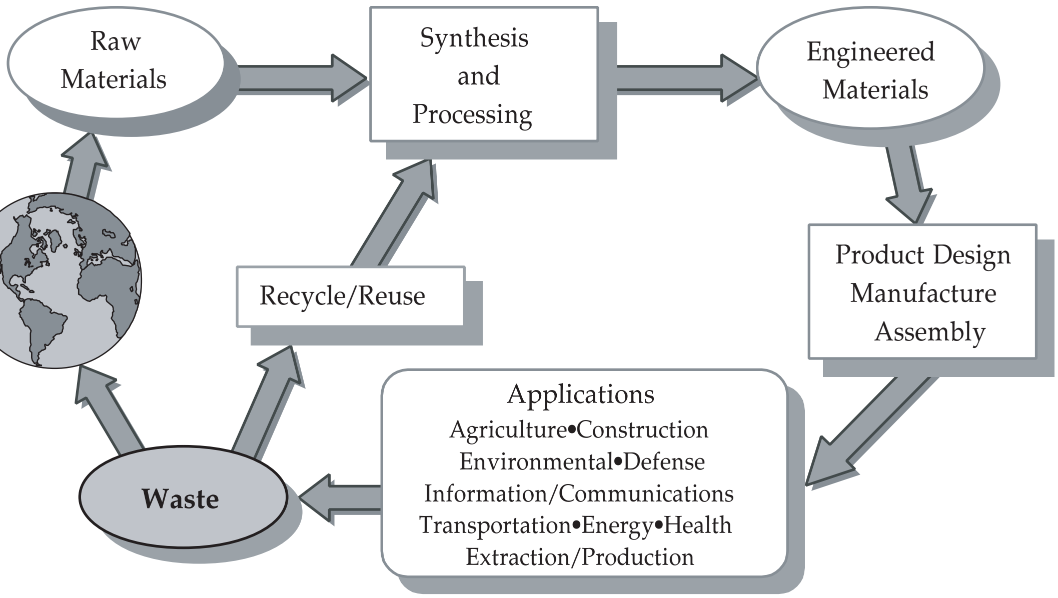


Figure 1. The global materials cycle

What kind of wastes can be vitrified?

Development of "cradle-to-grave" vitrification processes have been investigated and initiated by SRTC for wastes, which include, but are not limited to, the following:

- Spent filter aids from waste water treatment
- Waste sludges and liquid supernates including EPA hazardous sludges from harbors
- Mining industry wastes, sludges, and mill tailings
- Incinerator ash, incinerator offgas blowdown, or combinations of the two
- Lead paint
- Cement formulations in need of remediation
- Ion exchange resins and zeolites
- Soils, geologic material, or media, including naturally occurring radioactive material (NORM)
- Asbestos containing material (ACM) or inorganic fiber filter media
- Radioactive materials, including transuranic (TRU), plutonium (Pu), and other actinide wastes (e.g., Am and Cm)

Mixed low-level waste (MLLW)[4] in any of the above categories must meet the regulatory requirements imposed on hazardous waste by the EPA Resource Conservation and Recovery Act (RCRA) and the regulatory requirements imposed on radioactive materials governed by the U.S. Department of Energy (DOE) orders or Atomic Energy Act (AEA) regulations. Untreated wastes that fail the U.S. Environmental Protection Agency (EPA) Characteristically Toxicity Hazardous Leaching Procedure (TCLP) for any of the inorganic species listed in Table 1 (Column A) or any organic species listed in the Resource Conservation and Recovery Act are considered characteristically hazardous.[4] Prior to May 28, 1998, characteristically hazardous wastes could be treated so that they would meet the TCLP leachate levels given in Column A. The U.S. EPA promulgated a regulation on May 28, 1998, that characteristically hazardous wastes must be treated to the Universal Treatment Standards (UTS)[5] shown in Table 1 (Column B). The final vitrified glass must not release any of the species listed in Table 1 above the limits given in Column B whether the waste is non-radioactive or radioactive.

Table 1. Environmental Protection Agency Concentration Limits for Inorganic Constituents of Hazardous Wastes

	DWPF HLLW Sludges	SRS M-Area Sludges	SRS CIF Ash	SRS Am/Cm Solutions	SRS Asbestos Covered Pipe
Analyze Waste	1975	1987	1/92	1996	6/94
Surrogate Proof of Principle		1987	5/92	1996	4/96
Actual Waste Proof of Principle		1993	❋	O	4/98
Surrogate Pilot-Scale Testing		12/93	2/94	N/A	❋
Actual Waste Pilot-Scale Testing		9/94	❋	N/A	❋
Production Integrated Testing	3/95	V (4/96)	❋	1999	❋
Waste Processing	3/96	V (10/96)	❋	❋	❋
Recycle or Disposal	Disposal	Disposal	Disposal	Recycle	Recycle

** not underlying hazardous constituents

† Se must be treated to the characteristic limit (1.0 mg/L) to be non-hazardous, although it may be land disposed as a hazardous waste if <5.7 mg/L TCLP.

The need to provide MLLW treatment has been driven by the RCRA Land Disposal Restrictions (LDR) that require the treatment of the existing MLLW stockpiles. As of 1992 the MLLW waste volumes were ~250,000 m^3 and projected to increase to 1,200,000 m^3 by 1997 (Berry 1994). A schedule for DOE to come into compliance with RCRA was mandated by the passage of the Federal Facilities Compliance Act (FFCAct) of 1992. Large volumes of MLLW must, therefore, be converted to a solid, stabilized wasteform for permanent disposal. Since vitrification vaporizes EPA hazardous organics into CO_2 and H_2O, the final wasteform quality is assessed using the EPA Characteristically Hazardous Leaching Procedure for the inorganic hazardous species listed in Table 1.

A total of 76% of the existing mixed wastes in the DOE complex are candidates for electric and/or Joule heated vitrification (Berry 1994). Several RCRA listed MLLW wastewater sludges at SRS (Jantzen et al. 1993a; Jantzen et al. 1993b; Jantzen et al. 1994) and ORR (Jantzen et al. 1995) were identified as the first candidates for demonstration of Joule-heated vitrification. Several radioactive simulated RCRA wastes have also been shown to be candidates for vitrification and include incinerator ash and blowdown from SRS (Jantzen et al. 1993a; Jantzen et al. 1993c), waste sludge from Rocky Flats admixed with Portland cement, sludge from Los Alamos National Laboratory, and mill tailings from the Fernald Environmental Management Project (FEMP) K-65 site. Non-radioactive RCRA wastes successfully made into glass include waste water treated sludges from mining operations in Colorado, Pb paint from the Triborough bridge in New York City, and New York City harbor sludge. All of these wastes were rendered non-hazardous by the vitrification treatment, and the waste product could be recycled rather than disposed of. Vitrification studies of non-RCRA wastes have also been initiated by SRS. These include both non-contaminated ("clean") as well as radioactively contaminated asbestos containing material (ACM) from the DOE complex (Jantzen patent pending; Jantzen and Pickett 2000), some ion-exchange resins from commercial and government nuclear reactors (Jantzen et al. 1995), recycle of SRS americium and curium wastes to ORR for medical applications (Ramsey et al. 1995; Ramsey et al. 1994; Fellinger et al. 1998a; Fellinger et al. 1998b; Marra et al. 1999a; Marra et al. 1999b; Peeler et al. 1999a; Peeler et al. 1999b; Peeler et al. 1999c), and vitrification of weapons-grade and scrap plutonium (Ramsey et al. 1995; Ramsey et al. 1994) from the DOE complex.

Why vitrify?

Vitrification of radioactive or hazardous wastes into glass is an attractive option because it atomistically bonds the hazardous and radioactive species in a solid glassy matrix. The wasteforms produced are, therefore, very durable and environmentally stable over long-time duration. The Environmental Protection Agency has declared vitrification the Best Demonstrated Available Technology (BDAT) for high-level radioactive waste (Federal Register 1990) and produced a Handbook of Vitrification Technologies for Treatment of Hazardous and Radioactive Waste (U.S. Environmental Protection Agency 1992).

Vitrification processes are flexible to process chemistry variations and can accommodate dry or wet wastes (e.g., the process is very robust). Vitrification is an ancient, well-established, and well-studied technology used in many commercial applications. A new generation of high throughput Joule-heated melters, available from the commercial glass industry, allow for rapid vitrification of large volumes of waste. These vitrification systems are compact enough to be transportable (e.g., the SRS Transportable Vitrification System [TVS]) (Whitehouse et al. 1995a; Whitehouse 1995b) (see Figure 2). This enables the Joule-heated melter to be transported from waste site to waste site. Induction melters with high throughput, also used in the commercial glass industry, are robust and compact enough to handle high throughput vitrification of TRU wastes in glovebox applications or canyon operations. Compact melter technology minimizes capital and operating costs, making vitrification cost-effective on a life-cycle basis compared to other stabilization technologies that do not support recycle uses (see Figure 3).

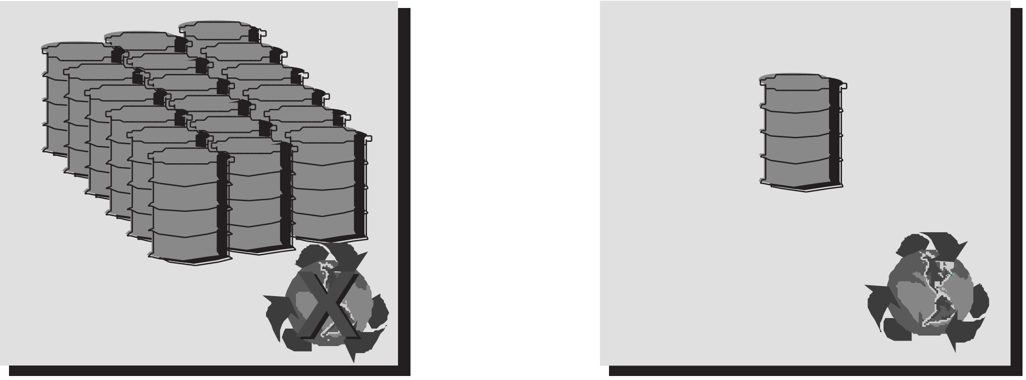
Vitrification produces large waste volume reductions (e.g., up to 97% [Jantzen et al. 1993a]) using cheap sources of glass former (e.g., sand, soil, crushed scrap fluorescent bulbs, crushed reagent bottles, etc.). Large reductions in volume minimize long-term storage or disposal costs if the waste cannot be recycled. Often the alternative stabilization technologies such as cement stabilization cannot produce a wasteform that is durable enough (e.g., cement does not thermally decompose the EPA RCRA hazardous organics and the porosity often allows the RCRA inorganic species to leach at greater than the UTS values listed in Table 1). Therefore, alternative stabilization technologies often cannot produce a wasteform that can be recycled (see Figure 3).

Developing a Vitrification Process

Development of each vitrification process follows the protocol shown in Figure 4 and below:

- Analyze wastes
- Surrogate proof-of-principle laboratory scale-studies (optional if actual waste is readily available)
- Actual waste proof-of-principle laboratory-scale studies

Figure 2. Transportable Vitrification System in the field. Composed of 5 modules, a feed batch preparation module (left) with an exterior waste tank or hopper, a melter module (center double story), an offgas module (along front face of concrete pad with a 40-foot tall offgas stack), a control/power supply module (behind the offgas and melter module). Co-designed by EnVitco Corporation and SRTC (U.S. Patent 5,611,766).



18 Drums (55 gallon) of cement vs. 1 Drums (55 gallon) of glass

1,440,000 (55 gallon) drums/year cement

7,930 (55 gallon) drums/year glass

Figure 3. Ninety four percent (94%) volume reduction for mining wastes vitrified at a conservative waste loading of 35 wt% compared to alternative stabilization in cement. Only 1 drum of glass, which can be recycled, instead of 18 drums of cement, which cannot be recycled.

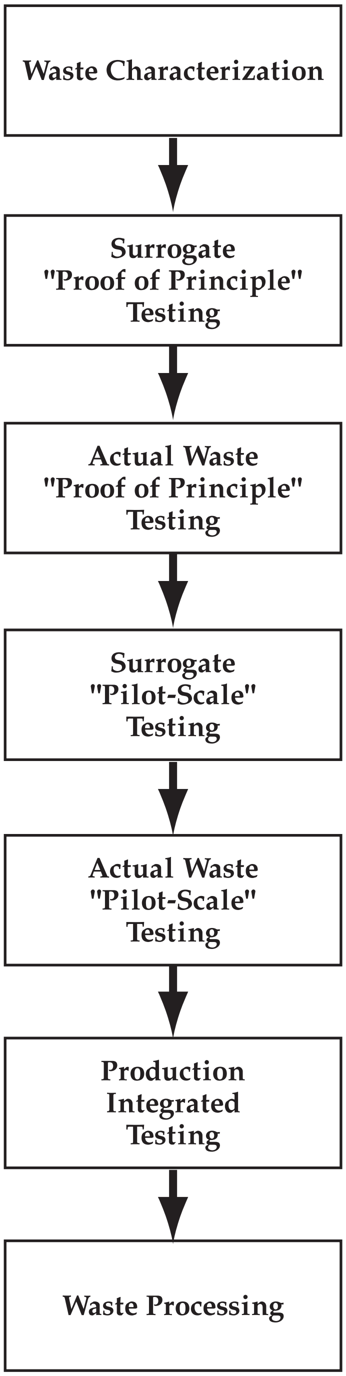


Figure 4. Steps in developing a vitrification process for any type of radioactive or hazardous waste. For some types of wastes certain steps are optional (see text).

- Surrogate pilot-scale demonstration (optional if actual waste is readily available)
- Actual waste pilot-scale demonstration
- Production scale (field-scale or full-scale) testing of melter with surrogate waste (necessary for initial check-out of equipment, otherwise optional)
- Actual waste processing (e.g., field-scale or full-scale)

The first step, proof-of-principle laboratory-scale testing, uses a systems approach to glass formulation and process optimization. The systems approach simultaneously evaluates product performance and processing considerations (Jantzen 1986; Jantzen 1991). Parameters affecting the product performance, such as chemical durability, are optimized relative to processing considerations such as melt temperature, volatility of hazardous species, melt viscosity, melt corrosivity, electrical resistivity, or waste solubility. The process or product models that form the basis for the statistical process control systems developed for HLLW (Jantzen and Brown 1993) and MLLW (Cozzi et al. 1999) vitrification, allow this optimization to be based on melter feed composition.

Proof-of-principle laboratory-scale crucible testing is often performed with surrogates to optimize glass product performance and processing considerations and if the amount of waste available is limited. Proof-of-principle laboratory-scale crucible studies are necessary with actual waste whether or not a surrogate is available. Proof-of-principle laboratory-scale crucible testing should evaluate the following parameters:

- Waste loading
- Melt temperatures
- Reduction/oxidation (redox) reactions between the waste and the additives
- Varying types of silica additives (e.g., Reactive Additive Stabilization Process [RASP])[6] using high surface area sources of silica such as various filter aids, perlite, precipitated silica,

rice husk ash vs. conventional vitrification with granular sand, soil, scrap glass from light bulbs reagent bottles
- Corrosion of melter materials of construction (refractories and electrodes)
- Determination of glass homogeneity (e.g., crystallization and/or phase separation)
- Wasteform performance (durability) evaluation using the Environmental Protection Agency Toxic Characteristic Leaching Procedure and/or the Product Consistency Test developed for HLLW and MLLW waste glass durability testing (ASTM C1285-97)
- Utility of existing statistical process/product control models (Jantzen and Brown 1993; Cozzi 1999)

Proof-of-scale-up testing is usually necessary in a pilot-scale melter. Pilot-scale testing with actual waste allows the following parameters, which cannot be assessed in crucible scale testing, to be determined:

- Data on actual vitrified wasteforms for input to Delisting Petitions for final disposal of listed wastes
- Confirmation of the processability of the glass compositions optimized in the proof-of-principle studies
- Determination of offgas emissions[7] as a function of melt temperature
- Verification of melter behavior as a Continuously Stirred Tank Reactor (CSTR) to ensure that waste and glass formers are homogenized during melting
- Demonstration of recycle of secondary waste condensate produced
- Utility of existing statistical process/product control models (Jantzen and Brown 1993; Cozzi 1999)
- Demonstration of decontamination of the offgas system/condensate tank
- Evaluation of melter refractory and electrode corrosion
- Determination of glass homogeneity (e.g., crystallization and/or phase separation)

The same steps were used to develop the vitrification process for HLLW vitrification at SRS and West Valley Fuel Services (WVFS). Although development of the process for vitrification of HLLW took ~25 years to develop, and the process for the M-Area waste sludges took ~7 years, the development of the ORR waste vitrification was completed in ~3 years. Tables 2-4 show the various vitrification projects initiated by SRS within the DOE complex and in the commercial sector. All of these vitrification projects were applications of the vitrification technology developed for HLLW disposal.

Successful Demonstrations of Waste Vitrification: Case Studies

RCRA Listed Radioactive Waste Sludges (Sometimes Admixed with Spent Filter Aids, Soils, and/or Cements)

SRS M-Area Sludge + Spent Filter Aid - 3,500,000 kg

- Analyze wastes (SRS) - high SiO_2 (~45 wt%), Al_2O_3 (~20 wt% as $Al(OH)_3$), $NaNO_3$ (~20 wt%) RCRA listed F006 nickel plating line sludge mixed with spent filter aid, Ni at ~1.2 wt% is the primary hazardous constituent, while the prime radioactive constituent is ~4.2 wt% U (Jantzen et al. 1993a).
- Actual waste proof-of-principle studies (SRS) - 44 glass formulations (alkali borosilicate[8] and alkali-lime-silica[8] glasses); waste loadings between 70-90 wt%; melt temperatures between 1150-1350°C; varied composition of waste from high alkali to high silica; 1 to 3 glass-forming additives; volume reductions of 86-88%; all glasses passed (Jantzen et al. 1993a; Jantzen et al. 1993b; Jantzen et al. 1994) the TCLP Land Disposal Restriction Universal Treatment Standards (LDR/UTS [Federal Register 1994]) in 1994, which were more stringent for Ni than the 1998 standards given in Table 1.

Table 2. Vitrification Projects Initiated by Savannah River Site at SRS

	DWPF HLLW Sludges[a]	SRS M-Area Sludges	SRS CIF[b] Ash	SRS Am/Cm Solutions	SRS Asbestos Covered Pipe
Analyze Waste	1975	1987	1/92	1996	6/94
Surrogate Proof of Principle		1987	5/92	1996	4/96
Actual Waste Proof of Principle		1993	❋	1996	4/98
Surrogate Pilot-Scale Testing		12/93	2/94	In Progress	❋
Actual Waste Pilot-Scale Testing		9/94	❋	N/A	❋
Production Integrated Testing	3/95	V (4/96)	❋	12/98	❋
Waste Processing	3/96	V (10/96)	❋	❋	❋
Recycle or Disposal	Disposal	Disposal	Disposal	Recycle	Recycle

Table 3. Vitrification Projects Initiated by Savannah River Site at/for Other DOE Sites

	ORR WETF[c] Sludges	ORR K-25 ±CNF[d] Sludges	ORR CPCF[e] Sludges	LANL[f] Sludges	RF Sludges	FEMP[g] Mill Tailings
Analyze Waste	1/94	1/94	7/95	1/94	1/94	1/93
Surrogate Proof of Principle	3/94	1/95	N/A	8/94	5/94	5/94
Actual Waste Proof of Principle	2/95	1/96	8/95	❋	❋	❋
Surrogate Pilot-Scale Testing	4/95	11/95	❋	9/94	6/94	❋
Actual Waste Pilot-Scale Testing	V	4/97	❋	❋	❋	❋
Production Integrated Testing	❋	10/96	❋	❋	❋	❋
Waste Processing	❋	6/97	❋	❋	❋	❋
Recycle or Disposal	Disposal	Disposal	Disposal	Disposal	Disposal	Disposal

■ Completed by SRS
■ Completed by other organization
V Vendor privatized
❋ Programmatic/budgetary hold

a DWPF HLLW - Defense Waste Processing High-Level Liquid Waste
b CIF - Consolidated Incinerator Facility
c WETF - West End Treatment Facility
d CNF - Central Neutralization Facility
e CPCF - Central Pollution Control Facility
f LANL - Los Alamos National Laboratory
g FEMD - Fernald Environmental Management Project

Table 4. Vitrification Projects Initiated by Savannah River Site at/for Commercial Firms

	Commercial Reactor Resins	Colorado Mining Industry	Pb Paint Removal by Thermal Spray Vitrification (TSV)	New York Harbor Sludge
Analyze Waste	4/96	5/98	1995	1995
Surrogate Proof of Principle	4/96	N/A	1995	N/A
Actual Waste Proof of Principle	❋	8/98	N/A	1996
Surrogate Pilot-Scale Testing	4/99	❋	N/A	N/A
Actual Waste Pilot-Scale Testing	❋	❋	1995	1997
Production Integrated Testing	❋	❋	N/A	❋
Waste Processing	❋	❋	❋	❋
Recycle or Disposal	Disposal	Recycle	Recycle	Recycle

■ Completed by SRS
■ Completed by other organization
V Vendor privatized
❋ Programmatic/budgetary hold

- Surrogate pilot-scale demonstration (SRS/Clemson DOE/Industry Waste Vitrification Center) - 6 sodium borosilicate glass formulations; one glass forming additive; waste loadings between 70-95 wt%; melt temperatures 1150-1500°C, all glasses passed TCLP LDR/UTS limits (Bennert et al. 1994).
- Actual waste pilot-scale demonstration (SRS) - 2 alkali borosilicate glass formulations[8]; waste loadings of 80 wt%; processed 400 kg of waste; all glasses passed TCLP LDR/UTS limits; TCLP and Multiple Extraction Procedure (MEP), which is multiple TCLP tests used for "Delisting" listed RCRA wastes; first Delisting Petition in the DOE complex for vitrified mixed waste (Poulous et al. 1995).
- Production/integrated full-scale testing (GTS Duratek) - first commercial vitrification of MLLW in DOE complex; contract awarded November 1993, design, construction, and a Readiness Review completed April 1996, simulant testing completed November 1996 (Pickett et al. 1994; Pickett and Norford 1999) (see Figure 5).
- Actual full-scale waste processing (Duratek) - fixed unit price treatment contract; all construction and operations costs borne by subcontractor until waste treated to meet delisting standards; treatment of M-Area wastes completed in February 1999 (Pickett et al. 1994; Pickett and Norford 1999).
- M-Area Vitrification Summary - first completed privatized vitrification project in the DOE complex. It was under budget, all proposed wastes were treated successfully, and the final glass met all product criteria (TCLP leaching on every batch). The volume of waste was reduced from >760,000 to >200,000 gallons (as glass "gems" in 71-gallon square drums), which will be delisted and

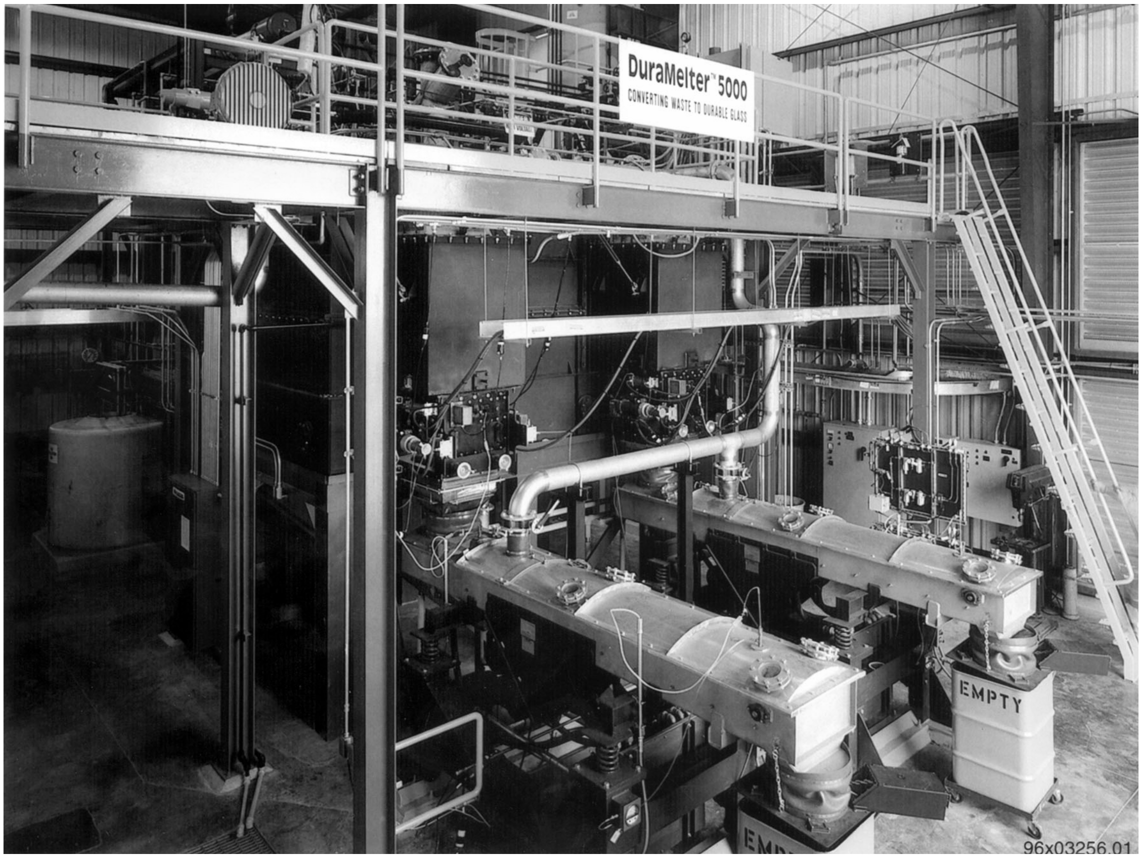


Figure 5. GTS Duratek Duramelter™ 5000 at the SRS M-Area Vitrifying RCRA Listed Waste Sludges from Ni Plating Line Operations. First vendor privatization of vitrification in the DOE complex.

disposed as low-level radioactive waste. From an SRS prospective, it was completely and totally successful privatization (Pickett and Norford 1999).

ORR West End Treatment Facility (WETF); ~8,000,000 kg

- Analyze wastes (ORR) - wastes contain 60-75 % CaO from $CaCO_3$, and 2-10 wt% Fe_2O_3 from FeOOH). RCRA listed waste from treatment of solvent residues plating line operations; nickel (~0.25 wt% is the primary hazardous species of concern while U at ~0.42 wt% and traces of Tc^{99} and TRU Np^{237}, etc.) are the radioactive species of concern (Bostick 1994).
- Surrogate waste proof-of-principle studies (SRS) - 120 alkali-lime-silica glass formulations; waste loadings of 20-70 wt%, melt temperatures between 1150-1350°C; no more than three glass-forming additives; severe melt line and general refractory corrosion at high waste loadings and high temperatures; sources of alkali and silica varied; glass viscosity vs temperature studied; all glasses passed TCLP LDR/UTS limits and PCT durability testing (Jantzen et al. 1995)
- Actual waste proof-of-principle studies (SRS/ORR) - ~60 alkali-lime-silica glass formulations with Tank 8 and Tank 13 due to the large known immiscibility gap in the $CaO-B_2O_3-SiO_2$ system (Volf 1984) where glasses are known to phase separate (form immiscible liquid phases); waste loadings between 20-70 wt%; melt temperatures between 1150-1350°C; no more than three glass-forming additives; volume reductions of 73-87%; sources of alkali

and silica varied; all glasses passed TCLP LDR/UTS.

- Surrogate pilot-scale demonstration (SRS/ Clemson DOE/Industry Waste Vitrification Center) - 2 alkali-lime-silica glass formulations; 3 glass-forming additives; waste loadings 20-40 wt%; melt temperature 1050-1350°C; 20 wt% glass passed TCLP LDR/UTS limits (Hewlett 1994); 40 wt% glass crystallized in the canister but passed TCLP.
- Actual waste pilot-scale demonstration - vendor privatized by ORR.

ORR K-25 B&C Pond Waste (Valley of the Drums); ~16,000,000 kg

- Analyze wastes (ORR) - B&C Pond Waste contained high SiO_2 (wt%) and CaO (~25 wt% from $Ca(OH)_2$) sludge with Fe_2O_3 (~16 wt%) from admixed clay basin liner, RCRA-listed mixed F006 wastes derived from plating line activities, Ag and Ni (~0.51 wt%) are primary hazardous components, ~0.30 wt% U is the primary radioactive constituent, trace concentrations of Tc^{99} (Bostick 1994); the relative proportions of SiO_2, $Ca(OH)_2$ and Fe_2O_3 vary greatly from drum to drum since clean RCRA closure of the basins in 1988-89 involved intermixing pond sludge with dredged clay pond liner and some partially successful stabilization efforts with Portland cement. The B&C Pond Waste was co-vitrified with CNF wastes containing high P_2O_5, high CaF_2, and high Fe_2O_3 (see discussion of CNF wastes below).
- Surrogate waste proof-of-principle studies (SRS) - 120 alkali-lime-silica glasses made with waste extremes; waste loadings of 40-90 wt%, melt temperatures between 1150-1350°C; a maximum of 3 glass-forming additives; sources of alkali and silica varied; general refractory corrosion studied, PO_4 solubility studied, glass viscosity vs. composition examined; all glasses passed TCLP LDR/UTS limits and PCT durability testing; crystallization and liquidus vs. composition studied.
- Actual waste proof-of-principle studies (SRS/ ORR) - 70 alkali-lime-silica glass formulations with waste from the rotary drier used in K25 B/C pond remediation efforts in 1991-92; waste loadings between 40-90 wt%; melt temperatures between 1150-1350°C; no more than 3 glass-forming additives; volume reductions of 70-90%; sources of alkali and silica varied; all glasses passed TCLP LDR/UTS.
- Surrogate pilot-scale demonstration (SRS/ Clemson DOE/Industry Waste Vitrification Center) - high SiO_2 B/C simulant developed by SRS; 1 alkali-lime-silica glass; three glass forming additives; waste loading 50 wt%; melt temperature 1250°C; glass passed TCLP LDR/ UTS limits and PCT testing
- Actual waste pilot-scale demonstration (SRS/ CETL) - at the Clemson Environmental Technologies Lab (CETL) during May and June of 1997. Melted two different waste streams: surrogate B&C pond waste and a blend of surrogate B&C pond waste with actual ORR Central Neutralization Facility (CNF) waste sludge (see discussion of CNF waste below). 865 kg of actual CNF waste sludge (see next section) was processed with a mix of B/C sludge making about 460 kg of glass
- Production/integrated field-scale testing (SRS) - high SiO_2 B/C simulant developed by SRS; SRS Transportable Vitrification System (TVS); waste loading 50 wt%; melt temperature 1150°C. An extensive surrogate waste test program was conducted on the TVS at Clemson during January and February of 1996. A total of 11,614 kg of surrogate waste glass was produced. Additional surrogate testing was performed at ORR in the fall of 1996.
- Actual field-scale waste processing (SRS) - The TVS treated a total of 7,345 kg of actual mixed waste composed of B&C pond waste and CNF waste. During the campaign at ORR's East Tennessee Technology Park (formerly ORR's K-25 site) 3,797 kg of B/C sludge and 3532 kg of CNF sludge were co-vitrified producing 7,970 kg of mixed waste glass during September and October of 1997 (Cozzi et al. 1999). Air pollution emissions did not exceed authorized limits and the glasses produced easily passed TCLP limits. The estimated volume reduction was 60%.

ORR Central Pollution Control Facility (CPCF); ~186,200 kg

- Analyze wastes (ORR) - there are three categories of CPCF wastes: oily, wet non-oily, and dry non-oily; oily RCRA listed plating line sludges containing 20-30% organics and ~0.50 wt% U and 0.2 wt% Ni. The oily CPCF wastes studied are high in SiO_2 (~50 % on a dry oxide basis), ~4 wt% CaO as $Ca(OH)_2$, ~12 wt% Fe_2O_3 from FeOOH, and ~30-40 wt% organics.
- Actual waste proof-of-principle studies (SRS/ORR) - 30 alkali-lime-silica glasses tested with oily CPCF waste and 3 glass formulations in the alkali-borosilicate system; waste loadings between 70-90 wt%; melt temperatures between 1150-1350°C; 3 glass-forming additives; volume reductions of 85-90% sources of alkali and silica varied; organics driven off with slow heat up ramps; all alkali-lime-silica glasses passed TCLP LDR/UTS; 3 borosilicate glass formulations phase separated (Pickett and Norford 1999) and had poorer overall durability.
- Actual waste pilot-scale demonstration (SRS/CETL) – not completed because a pretreatment technique such as solvent extraction, wet oxidation, or incineration was needed to destroy the 30-40% organics before vitrification (the maximum safe organic content for a Joule heated melter is <10 wt% organics).

ORR Central Neutralization Facility (CNF); ~900,000 kg

- Analyze wastes (ORR) - CNF wastes are listed RCRA wastes resulting primarily from the treatment of ORR TSCA incinerator scrubber blowdown solution.
- Surrogate waste proof-of-principle studies (SRS) - 15 alkali-lime-silica glass compositions tested with waste loadings ranging from 15 to 40 wt%; melt temperature was 1250°C; glasses with higher Li_2O content produced more homogeneous glasses.
- Actual waste proof-of-principle studies (SRS/Clemson) – 6 glass compositions tested in the ALS system with waste loadings ranging from 30 to 40 wt%; melt temperature was 1250°C; glasses were visually homogeneous.
- Actual waste pilot-scale demonstration (SRS/CETL) - co-vitrified with B&C pond waste (see previous section).
- Actual field-scale waste processing (SRS) - performed as part of the TVS campaign on B&C pond waste in 1997. See discussion above.

Los Alamos National Laboratory (LANL) Liquid Waste Processing Plant; ~ 324,000 kg

- Analyze wastes (ORR) - ~50 wt% CaO (on an oxide basis) from $CaCO_3$ processing and admixed Portland cement and gypsum, high SiO_2 (38 wt% from filter aids such as perlite and diatomaceous earth), and Fe_2O_3 (8 wt% from FeOOH) RCRA from treatment of solvent residues; U at ~0.23 wt% and traces of Pu^{239} and Am^{243} are the radioactive species of concern and the hazardous species of concern are not well documented except for Cd (Bostick 1994).
- Surrogate waste proof-of-principle studies (SRS) - 19 alkali borosilicate glasses were tested, as well as glasses in the $CaO-Al_2O_3-SiO_2$, the $CaO-Fe_2O_3-SiO_2$, and the soda-lime-silica glass (SLS) forming systems (Cicero et al. 1995); waste loadings of 25-75 wt%, melt temperatures between 1150-1500°C; 2 glass-forming additives; severe crystallization was noted in certain composition regions in all systems with the in the SLS glasses. Glasses doped with Ba, Cd, Cr and Ni; all glasses passed TCLP LDR/UTS limits and PCT durability testing (Cicero et al. 1995).
- Surrogate "pilot-scale" demonstration (SRS/Clemson DOE/Industry Waste Vitrification Center) - 1 glass formulation at 65 wt% loading in the $CaO-Al_2O_3-SiO_2$ system at 1350°C, the glass was difficult to pour due to high viscosity, TCLP, and PCT testing indicated durable glass was produced.

Rocky Flats By-Pass Sludge

- Analyze wastes (ORR) - ~36 wt% Fe_2O_3 from $Fe(OH)_3$, ~25 wt% CaO from $CaSO_4$, and ~12 wt% Na_2O from $NaNO_3$ creating about 12 wt% NO_x and >20 wt% SO_3 gaseous species upon vitrification; some waste admixed with up to 30% Portland cement; RCRA listed nickel plating line waste; listed for Cd, Cr, Pb, Ag, and Ni hazardous species; Pu as primary radioactive species of concern (Bostick 1994)

- Surrogate waste proof-of-principle studies (SRS) - 10 sodium-borosilicate glass formulations were tested at waste loadings of 25–75 wt% but required charcoal additions to get rid of the sulfate layer that formed on the glass surface. Only 2 glass-forming additives plus charcoal were necessary to stabilize the waste. The homogeneous glasses passed TCLP LDR/UTS limits and PCT durability testing (Cicero et al. 1995).
- Surrogate "pilot-scale" demonstration (SRS/Clemson DOE/Industry Waste Vitrification Center) - 1 glass formulation at 75 wt% loading in the sodium-borosilicate system at a melt temperature of 1350°C.

Incinerator Wastes (Ash and/or Offgas Blowdown)

SRS Consolidated Incinerator Facility (CIF); ~ 800 m^3/year blowdown and 124 m^3/year ash generation for 25+ years

- Simulate wastes (SRS) - ~65 wt% Na_2O (primarily from NaCl in the waste) and ~32 wt% CaO (on a dry oxide basis) in a mixture of 68 wt% blowdown and 32 wt% bottom ash (Jantzen et al. 1993a); RCRA for all inorganic species of concern and Zn; radioactive constituents include Cr^{51}, Sr^{90}, Cs^{137}, traces of Pu.
- Surrogate waste proof-of-principle studies (SRS) - 20 alkali-lime-silica glasses tested with surrogates (Jantzen et al. 1993a); waste loadings of 30-50 wt%; melt temperatures between 1150-1250°C to avoid volatilization of hazardous species such as chlorides; 94-97% volume reduction; 1 glass forming additive, SiO_2; sources of alkali and silica varied; all glasses passed TCLP LDR/UTS limits; pyrohydrolysis investigated to remove Cl as HCl plus steam (Jantzen et al. 1993a)

Ion Exchange Resins and Zeolites

Commercial Reactor Resins ~220,000 kg Boiling Water Reactor (BWR) and 66,000 kg Pressurized Water Reactor (PWR) Ion Exchange (IEX) resins per reactor per year

- Analyze wastes (SRS/EPRI) - samples of 6 ion exchange resins from EPRI undergoing wet chemical analysis for cationic and anionic species; undergoing Differential Thermal Analysis (DTA) with coupled mass spectrometry to identify inorganic and organic volatile components
- Surrogate waste proof-of-principle studies (SRS) - preliminary data indicates 50 wt% waste loading for PWR wastes which gives a 77% volume reduction and a 35 wt% waste loading for BWR wastes, which gives a 66% volume reduction (Jantzen et al. 1995); 35 glass compositions tested with 6 different resin types with a borosilicate glass composition; waste loading (24 to 42 wt%) limited by salt formation and glass redox; melt temperature of 1150°C; glasses passed PCT durability testing.
- Surrogate pilot-scale demonstration (SRS)/Clemson) - 1 borosilicate glass formulation tested at 33 wt% waste loading for a 64% volume reduction; melt temperature of 1050°C; glass was homogeneous and passed PCT durability testing; retention of radioactive surrogates was greater than 93% (Cicero-Herman et al. 1999).

National Atomic Energy Commission of Argentina (CNEA) Resins – 42 m^3 with 2.83 m^3 generated per year of Atucha and 130 m^3 with 9.5 m^3 generated per year of Embalse

- Analyze wastes (SRS/CNEA) - resins analyzed by the CNEA; surrogate radionuclides added by SRS and the CNEA for vitrification testing.
- Surrogate waste proof-of-principle studies (SRS) – a borosilicate glass composition tested with both resins at various waste loadings; optimum determined to be ~30 wt% waste loading for both resins with associated vol-

ume reductions of >65 %; melt temperature of 1050°C; glasses were homogeneous and durable.
- Surrogate pilot-scale demonstration (SRS/Clemson) − 3 melter demonstrations completed with simulated and actual CNEA supplied resin; melt temperature of 1050°C; homogenous and durable glasses produced.

Soils, Geologic Material/Media

Fernald Environmental Management Project (FEMP) K-65 silos of depleted uranium (mill tailings from processing pitchblende ore) for atomic bomb development ~10,000,000 kg (~10,000 metric tons)

- Analyze wastes (Merrill and Janke 1993) - Residues from processing pitchblende ores from 1949-1958, high in SiO_2 (~63 wt%), BaO (~6.5 wt%), Pb (~12.5 wt%), and Fe (~5 wt%)
- Surrogate waste proof-of-principle studies (SRS) - 2 alkali-lime-silica glass formulations; waste loadings of 80-90 wt%, melt temperature 1050°C; two glass forming additives; both glasses passed TCLP limits (Jantzen et al. 1999)

Asbestos and/or Glass Fiber Filters (Uncontaminated or Contaminated)

Decommissioning and Decontamination (D&D) throughout the DOE, DOD, and commercial sectors (Jantzen and Pickett 2000)

- Analyze wastes (SRS) - analysis of asbestos coated pipe indicates that asbestos containing materials (ACM) are admixed with up to 50 wt% $MgCO_3$ and/or $CaSO_4$ as cementitious binder material
- Surrogate waste proof-of-principle studies (SRS) - use of patented (Jantzen pending) caustic dissolution process to remove ACM from adhering pipe; allows pipe or other adhering metal to be sold/recycled; 10 glass formulations of high Mg silicate glasses render ACM non-crystalline and non-hazardous; waste loadings of 60-70 wt%; melt temperatures between 1150-1350°C; volume reductions of 90-99.7% for asbestos covered pipe; non-contaminated glass can be sold for recycle.

Radioactive Materials Including Transuranic (Tru), Plutonium (Pu), and Other Actinide Wastes

Am/Cm - 15,000 liters to be stabilized in glass for shipment to ORR for reuse as medical target sources (Ramsey et al. 1994; Fellinger et al. 1998a; Fellinger et al. 1998b; Marra et al. 1999a; Marra et al. 1999b; Peeler et al. 1999a; Peeler et al. 1999b; Peeler et al. 1999c)

- Analyze wastes (SRS) - dilute 4N nitric acid solution containing approximately 10.1 kg Am and 2.7 kg Cm
- Surrogate waste proof-of-principle studies (SRS) - compositional variability studies have demonstrated the production of glasses with 30−47 wt% feed loadings, coupled with a lanthanide borosilicate based frit, that meet specific process and product performance specifications. The resulting glass form can be safely shipped to ORR as a solid for their Isotope Sales Program; SRS waste reclaimed as a source of revenue for DOE complex; full (100%) recovery of all rare earth oxides (including La_2O_3, CeO_2, Er_2O_3, Eu_2O_3, and Nd_2O_3) from glass demonstrated by nitric acid extraction; >90% volume reduction.
- Surrogate waste full scale demonstration (SRS) − Actinide and lanthanide oxalates precipitated from solution with oxalic acid and then washed to lower the nitric acid concentration; oxalate precipitate is then transferred to a Pt/Rh induction melter, which is preloaded with glass-making additives; the mixture is dried and heated to approximately 1450°C in the induction heated Cylindrical Induction Melter (CIM) (see Figure 6); glass is poured through a drain tube into a stainless steel cylinder for shipment.

RCRA Hazardous Mining Industry Wastes (Jantzen et al. 2000)

- Analyze wastes (SRS) - ~7 wt% Al_2O_3, ~7 wt% CaO, ~20 wt% (Fe_2O_3 + FeO), ~12wt % MnO, ~ 25 wt% ZnO, and ~8-9 wt% SiO_2; RCRA hazardous for CdO which only comprises 0.12 wt% of the waste.

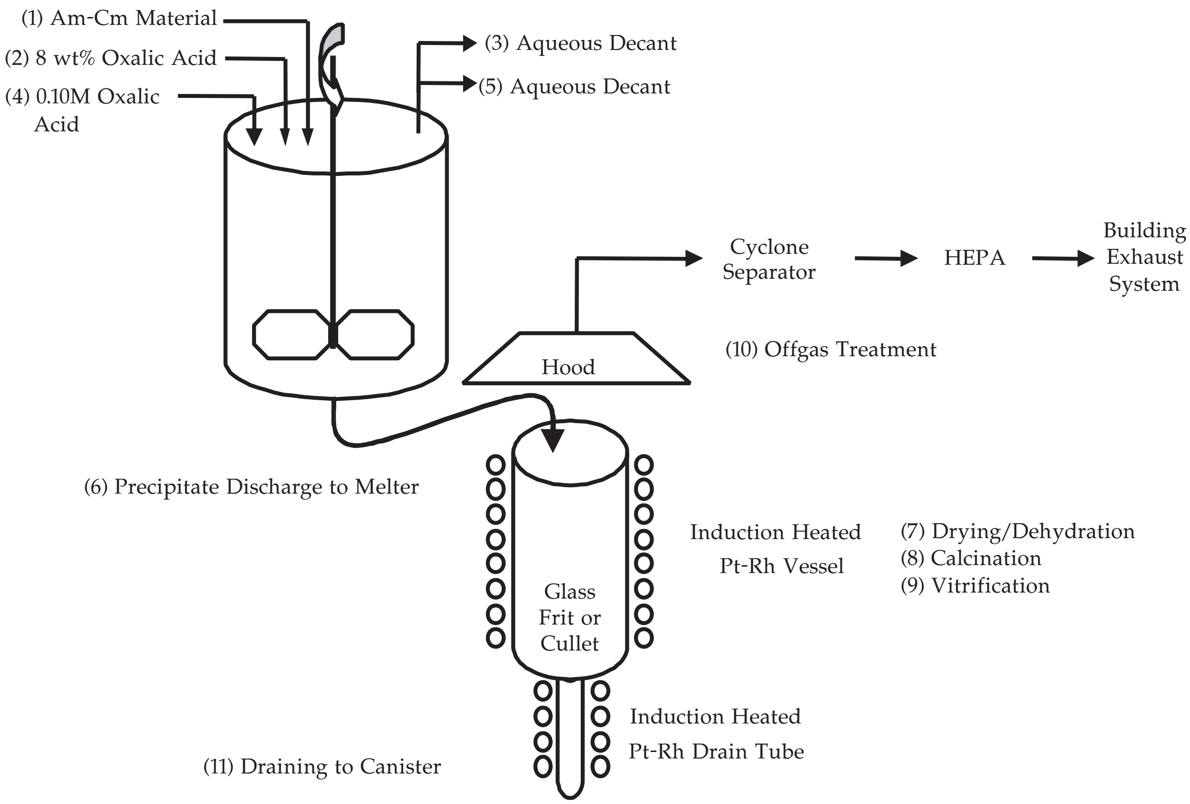


Figure 6. Schematic of the Batch Vitrification Process

- Surrogate waste proof-of-principle studies (SRS) - 9 soda-lime-silica glasses tested, waste loadings of 35-50 wt%, volume reductions of 90-94% (see Figure 3); melt temperatures of 1250-1350°C; 2 basalt glasses tested, waste loadings of 35 wt%; volume reductions of 90-94%, melt temperatures of 1300-1500°C; one borosilicate glass tested, waste loadings of 28 wt%, volume reductions of 87%, melt temperatures of 1150-1250°C; mill tailings from mine used as cheap source of glass forming additives; glasses rendered non-hazardous by TCLP and acceptable for recycle.

RCRA Hazardous New York Harbor Sludge (Marra 1996)

- Analyze wastes (WSTC) - Westinghouse Science and Technology Center ~60 wt% SiO_2 and ~15 wt% Al_2O_3; waste RCRA hazardous for organics such as dioxins and heavy metals such as Pb from fuel used in ships and boats
- Surrogate waste proof-of-principle studies (SRS) - 3 soda-lime-silica glasses tested, waste loadings of 85 wt%; melt temperature 1350°C; volume reductions of >90%; glasses rendered non-hazardous by TCLP and acceptable for recycle.

RCRA Hazardous Pb Paint Removal (Marra et al. 1996)

- Analyze wastes (SRS) - not applicable
- Surrogate waste proof-of-principle studies (SRS) - 4 alkali borosilicate, 1 sodium barium silicate, 1 lead iron phosphate, and 1 commercial lead silicate (leaded glass) glass formulas were tested; the borosilicate glasses containing lithium oxide were the most successful in stabilizing the hazardous Pb constituents.
- Actual waste full scale demonstration (USACERL) - the U.S. Army Corps of Engineers Construction Engineering Research Laboratory performed a full-scale demonstra-

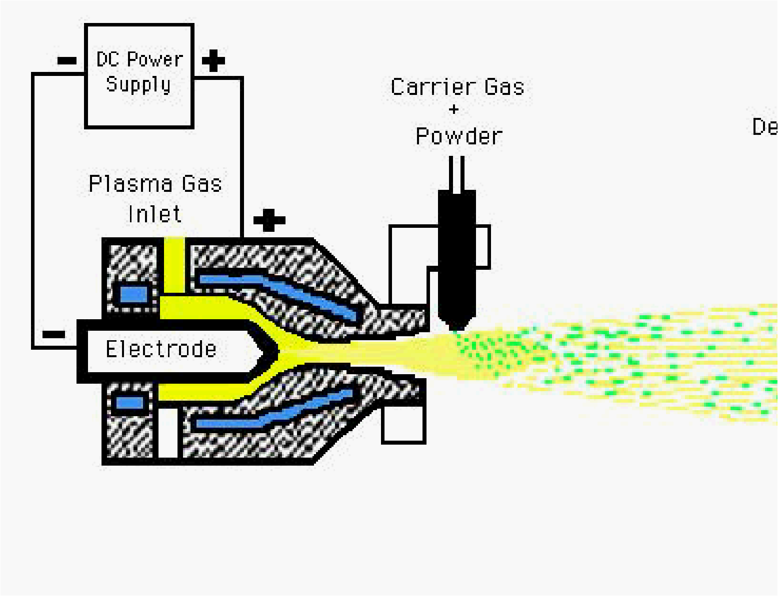


Figure 7. In thermal spray vitrification a high-temperature plasma carries a mixture of crushed glass powder and a carrier gas in a hot flame. The molten glass impinges on the contaminated or painted metal substrate. The high temperature vaporizes the organics in the paint and atomistically bonds the hazardous species (Pb in the case of paint wastes) in the glass. As the glass cools the thermal mismatch between the glass and the metal substrate causes the glass to crack off the substrate. The glass can then be swept up or vacuumed up for disposal or recycle.

tion of the Thermal Spray Vitrification (TSV) on the Triborough Bridge in New York City; removed multiple layers of paint that had accumulated on the bridge abutment over a 30 year period of time (see Figure 7).

Conclusions

Vitrification is a viable option for a large variety of wastes in the DOE complex and the commercial sector. SRS has initiated and completed vitrification projects in both stationary and transportable vitrification facilities as well as via thermal spray vitrification. A wide variety of waste types can be stabilized or recycled with this robust technology. In addition to the case studies presented in this review, SRS has recently initiated vitrification programs with the Idaho National Engineering and Environmental Laboratory (INEEL) for vitrification of their HLW calcines and with British Nuclear Fuel Ltd. (BNFL) for vitrification of HLW and MLLW Hanford wastes. One of the most recent endeavors has been the adaptation of TSV for decontamination of radioactive species from duct work and hoods throughout the DOE complex.

Endnotes:

1. The history of the development of the vitrification of HLLW at SRS is the subject of another section of this proceedings (Randall and Marra 2000). To date over 3 million pounds of HLLW waste glass have been produced in the SRS Defense Waste Processing Facility (DWPF).
2. Wastes that are both hazardous under the Environmental Protection Agency (EPA) Resource, Energy and Recovery Act (RCRA) and radioactive (e.g. governed by the Atomic Energy Act [AEA]).
3. Joule-heated or induction-heated melters. Joule-heated melters vitrify waste in a refractory-lined vessel containing diametrically opposed electrodes. The electrodes are

used to heat the glass by passing an electric current through the material. The process is called Joule heating.

4. Waste that contains source, special nuclear, or byproduct material subject to regulation under the Atomic Energy Act and hazardous waste species subject to regulation under the Resource Conservation and Recovery Act waste as defined in 40 CFR 261 (U.S. Code Title 42, Section 2011)
5. Federal Register, V.63, #100, p. 28748-9.
6. Reactive Additive Stabilization Process (RASP), U.S. Patent 5, 434,333. Reactive high surface area silica, used as a waste form additive, was determined to greatly enhance the solubility and retention of hazardous, mixed and heavy metal species in glass (Jantzen et al. 1994; Jantzen 1995). Vitrification using this Reactive Additive Stabilization Process (RASP)* was found to increase the solubility and tolerance of Soda (Na_2O)-Lime (CaO)-Silica (SiO_2) glass (SLS) to atomistically bond waste species. Highly reactive silica lowers glassification temperatures; increases waste loadings, which provides for large waste volume reductions; minimizes melt line corrosion; and produces EPA acceptable glasses.
7. The EPA, as part of the Clean Air Act, Title III, National Emissions Standards for Hazardous Air Pollutants (NESHAP), has imposed Maximum Achievable Control Technology (MACT) standards on hazardous species such as Cr, Pb, Cd, Hg, Be, and As and a host of organics from all types of high-temperature thermal treatments from incineration to vitrification to cement kiln operations.
8. Soda-lime-silica glass is common window glass. Lithium oxide was used preferentially over sodium as a glass-forming flux additive and various silica sources were investigated since recent studies had shown that the known glass-forming region in the SLS system could be expanded using reactive sources of SiO_2 and or reactive fluxes like Li_2O ([Jantzen et al. 1993a; Jantzen et al. 1993b; Jantzen et al. 1994} U.S. Patent 5,434,333, Lithia Additive Stabilization Process, LAMP™, patent pending).

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Excellence in Control of Radiation Exposures

Kenneth W. Crase

Abstract

Savannah River Site (SRS) operating contractors have continuously maintained excellence in the control of radiation exposures to both Site workers and the public in the surrounding area. In doing so, Site organizations and radiation protection officials have contributed significant advances to the nuclear industry in methods for controlling radiation exposures. These methods, developed and first used at SRS, are now routinely used throughout the U.S. nuclear industry. As a consequence of their use, the average lifetime occupational radiation dose to workers monitored at SRS is among the lowest of major U. S. nuclear facilities. The average annual radiation dose to any monitored worker has been maintained as a small fraction of federal criteria. Public doses have long been maintained at less than a few percent of federal limits and much, much less than the average citizen receives from natural radioactivity.

Introduction

SRS is one the largest nuclear sites in the country. Due to the primary mission of nuclear materials production for national defense programs through much of its history, a significant fraction of the nation's inventory of radionuclides has been generated, processed, or stored at the Site. Without stringent controls for radiation exposures, the occupational and environmental legacy of SRS could have been adverse to worker and public health, as has been witnessed in similar facilities in the former U.S.S.R. Fortunately, the founding team that started up SRS recognized this potential and ensured appropriate and conservative radiation exposure control methods were developed and used at SRS to avoid such negative worker health and environmental legacies.

During the course of establishing and maintaining one of the largest U.S. radiation protection programs, there have been numerous technical contributions in several areas. Such developments include dosimeters to measure routine radiation exposure, accident dosimeters, equipment to measure dosimeter response to radiation, instruments to monitor radiation levels or presence of radioactive material, and protective equipment such as plastic suits for work in airborne radioactivity areas. Such instruments and protective gear had to be developed onsite initially because no commercial source was available. These technical contributions, though important to the radiation protection program at SRS and throughout the nuclear community, are not the focus of this report, and have been described in other reports (Taylor et al. 1995). Rather, the purpose of this paper is to describe techniques introduced at SRS over its history to control radiation exposures, and by so doing, pay tribute to the many SRS managers and staff who contributed to development of these methods. It is regrettable that it is not possible to specifically mention all such contributors.

Worker Radiation Exposure Experience

Land clearing for facilities construction at SRS began in 1951, with the first reactor (R Reactor) becoming operational in December 1953. Other major radiological facilities became fully operational over the next three years. Site facilities included a total of five heavy-water production reactors, two chemical separations facilities, fuel fabrications facilities, tritium extraction facilities, two plutonium production facilities, waste management facilities, supporting facilities and analytical laboratories, and applied research and development laboratories. Hazards included the full gamut of fission and activation products as well as uranium and transuranic radioisotopes such as plutonium. Thus, the radiation protection program needed to be able to address containment of these radioactive materials, to control radiation exposure of workers who worked with these materials, and to minimize radiation releases to the environment and public exposures.

Radiological work at SRS has resulted in a cumulative radiation dose of approximately 65,000 rem to about 70,000 workers monitored for radiation exposure. Thus the average lifetime radiation dose to a monitored worker is about 1 rem, which is among the lowest for the major Department of Energy sites. Naturally, the largest collective doses were delivered during years of greatest production and have declined significantly over recent decades. The SRS annual collective worker dose has ranged from a high of slightly over 3,000 rem in 1960 to the current level of about 100 rem.

Approximately 84% of the SRS cumulative occupational radiation dose to workers is due to external dose received from beta or gamma radiation, and it has been monitored by dosimeters worn on the torso of the body. Nearly 6% of the cumulative worker dose is due to neutron exposure, also monitored by dosimeters worn on the body. Tritium taken into the body accounts for about 4% of the cumulative dose. Tritium dose is monitored by urine analyses. Intakes of radionuclides other than tritium account for about 6% of the SRS cumulative worker dose. These intakes must be determined through a variety of analyses, such as whole body or chest counting, urine or fecal analyses, and the use of metabolic models to assess intake amounts and dose.

The average SRS worker monitored for radiation exposure has received an annual radiation dose that has varied from a maximum of 0.5 to 0.6 rem per year in the 1960s to a current low of about 0.015 rem per year.

Worker Exposure Control Methods

Radiation protection officials had the benefit of experience from less than a decade of operations of nuclear facilities built during the Manhattan Project. There were no academic programs at that time specifically for radiation protection, and no readily available pool of highly experienced health physicists. Initial radiation protection staff experience was achieved through hiring staff from existing nuclear facilities, particularly those at Hanford and Oak Ridge, but only a few could be hired. Much of the success of the SRS radiation protection program is due to the ingenuity and diligence of the early management and technical staff. The program also benefited by great stability. For example, the first manager of the radiation protection program, C. M. “Pat” Patterson, who transferred to SRS from Du Pont Hanford operations, managed the program continuously until his retirement in 1978. W. C. “Bill” Reinig (one of the very first exempt employees at SRS in June 1951) then managed the radiation protection program for more than a decade.

Administrative Dose Control Levels

At the time SRS was starting up in the early 1950s, there were no national limits for worker or public radiation dose. There was guidance from various organizations that recommended worker dose be maintained below what would now be the equivalent of about 15 rem per year. As early as 1949, however, guidance as low as 5 rem per year had been discussed in a meeting including representatives from the United States, Britain, France, and Canada. Subsequently, when the Federal Radiation Council was established in 1958, a 5-rem-per-year federal guidance was developed. The initial radiation protection team instituted the policy that SRS workers would be restricted to receive no more than 3 rem per year from whole body radiation dose (this includes beta, gamma, neutron, and tritium dose). This practice was due to conservatism toward worker safety and health and concerns over adequate means to monitor neutron exposures. It is the first known application of an administrative dose control level at a nuclear site, at levels more conservative than federal guidance. This administrative control level remained in effect through 1992. Since 1992, DOE has established a DOE administrative control level of 2 rem per year for workers in the DOE complex and has ordered that each DOE site establish an administrative control level. The SRS administrative control level was lowered to 1.5 rem per year for 1993 and has been lowered since to a current level of 0.5 rem per year for most SRS workers.

The early and continuous use of an administrative control level has been very effective at controlling individual and collective SRS worker doses. Very few individuals have ever exceeded the control level throughout the history of the Site, and each who has exceeded the control level was involved in a radiological incident where unplanned exposures or intakes of radionuclides occurred.

Radiological Work Permits

SRS at start-up instituted the development and use of Special Work Permits, which were required for radiological work where significant radiological hazards were present and where significant potential existed for larger exposures or for the spread of contamination. Later, such work permits, now generally called Radiological Work Permits, became common in the commercial nuclear power industry, and are now widely used throughout the U. S. nuclear industry. These permits include the level of hazard, such as dose rates available; the required protective gear and dosimetric requirements for the work; and a requirement for each person who enters to work in that area read, understand, and sign the permit.

Radiological Control Manual

SRS was one of the first nuclear facilities to develop and use a Radiological Control Manual (at SRS it was SRP DPSOP 40, *Radiological Controls*). This manual contained the rules for performing radiological work at SRS. Further, from very early in the Site's history, specific radiological safety procedures were instituted, called DPSOLs, which specified how the radiation protection program practices were performed. The early use of a radiological control manual and specific procedures for performing radiological work have been instrumental in controlling radiation exposures. Such practices are now used throughout the nuclear industry.

Dose-based Performance Goals

Starting in the early 1970s as the concepts of As Low As Reasonably Achievable (ALARA) were being developed by guidance agencies, SRS began using dose-based performance goals for maximum individual dose and for group collective dose. These goals have been aggressively pursued and lowered as Site operations permitted. For example, while the SRS Administrative Control Level was 3 rem per year for 1992, the goal was to have no worker receive more than 1.5 rem that year, and no one did. Goals for individual operating organizations have also been established. In addition to exposure performance goals, other radiological performance indicators have been added to this program over time.

ESH Review Committees

A significant contributor to the success of the ALARA goal process at SRS has been the establishment of a clear environment, safety and health management review process. Since such goals have been in place, a management safety committee chaired by the top contractor manager has periodically reviewed (monthly for most of this time) the radiological performance against goals. This committee, long called the

Site Central Safety Committee, reviews and provides focus on areas where improvement is needed or where special performance situations exist. A similar upper management committee approves goals set for the next year. Subcommittees for operating divisions contribute to the review and goal establishment process, and generate programs that enable improvement. The management review process has ensured that goals are aggressively pursued, and that goals for subsequent years are challenging.

Public Dose Experience

SRS has delivered a collective public radiation dose of about 5,000 rem to about 600,000 members of the public since site start-up (Carlton 1998). The maximum dose to any member of the public (the fence-line dose) was largest in the 1960s, when that dose was in the range of 0.1 to 0.5 rem per year. By the 1980s, the maximum public dose was in the range of 0.001 to 0.003 rem per year. Now, the maximum public dose is typically less than 0.0002 rem per year. The current federal limit for such exposures is 0.1 rem per year.

Methods for Control of Public Dose

Like the control of worker dose, SRS has developed methods to reduce the impact of Site operations on the environment and the public. As the Site was being cleared for construction of nuclear facilities, the radiation protection staff performed the first ever base-line study of environmental radiological conditions prior to construction and operation of Site facilities. This report has been highly useful in determining radioactivity concentrations in environmental media that is due to site operations as opposed to natural radioactivity (Reinig 1953).

In 1970, SRS established a dose-based technical standard for radioactivity releases from SRS facilities. This technical standard established that "we can and will operate this site in such a manner that no member of the public receives as much as 10 mrem in a year from site operations." Since establishment of this standard, it has always been met. In late 1982 and early 1983, as the Environmental Protection Agency was establishing numeric criteria for the Clean Air Act for radioactive releases, they adopted the SRS standard, and it remains the Clean Air Act atmospheric radiological release criteria for a nuclear facility to this day.

In addition to the dose-based overall atmospheric radiological release criteria, SRS also instituted in the 1970s specific atmospheric and liquid radiological concentration goals for specific release points, such as a facility stack or liquid release pipe into a site stream. Like the worker dose ALARA goals, these goals were aggressively and routinely reviewed and updated annually in a similar upper-management committee review process. The review process encouraged organizations operating radiological facilities to develop administrative and engineered improvements in their waste streams to meet ever more challenging goals. In recent years, these concentration goals have been replaced with dose fraction goals.

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Biography

Kenneth W. Crase is technical advisor to the Health Physics Technology organization of the Westinghouse Savannah River Company. Mr. Crase came to the Savannah River Site in July 1981. He has managed the environmental monitoring program and held various technical positions. He provides technical guidance to the SRS radiation protection program for workers and the environment.

He is a member of the Department of Energy Oversight Board for External Dosimetry for their Laboratory Accreditation Program, is an adjunct professor in the Nuclear Engineering and Health Physics Department of Georgia Institute of Technology and has taught graduate health physics courses via tele-lecture.

Mr. Crase received his Ph.D. in physics in 1971 at The University of Tennessee. He received his B. A. in physics at Berea College in 1966. He has previous radiation protection experience at Oak Ridge National Laboratory and at Lawrence Livermore National Laboratory and has taught college for four years.

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Advances in External Dosimetry at the Savannah River Site

Dante' W. Wells

Abstract

External dosimetry is the science of determining the external radiation dose to personnel that work in radiation fields. The determination of dose to the worker is required for beta, gamma, and neutron radiation that personnel are exposed to in the workplace. The Savannah River Site (SRS) has measured the external exposure to radiation workers since November 1951. During this nearly 50-year period, external radiation dosimetry methods and technological advancements have led to many improvements in techniques that are now used to quantify the measured occupational radiation dose. This paper presents the evolution of beta, gamma, and neutron external radiation dosimetry at SRS. This paper will also highlight the accomplishments of the talented personnel responsible for the innovative techniques created over the years to ensure the quality and validity of the external dosimetry results.[1]

Introduction

The science of external dosimetry was developed to determine radiation dose imparted to living tissue by three types of ionizing particles—beta radiation, gamma radiation, and neutron radiation. The region of the body affected by the action of a single ionizing particle is small and the damage caused is insignificant. However, the damage produced by successive particles accumulates, and, if enough energy is imparted, the consequences can become detrimental. External dosimetry programs determine the amount of dose accumulated over an extended period of time (i.e., monthly or quarterly). The Savannah River Site (SRS) has used several types of nuclear emulsion dosimeters and thermoluminescent dosimeters (TLDs) to maintain an increasingly cost-effective, timely, and accurate external dosimetry program.

Beta-Gamma Dosimetry, Film Badge Dosimeter (1951 to 1970)

Beta and gamma radiation monitoring at SRS began on November 12, 1951, with the distribution of 50 Oak Ridge National Lab (ORNL)-designed film badge dosimeters attached to the Site security badge. This original dosimeter used the Du Pont manufactured Type 552 x-ray film; however, a new film packet, Du Pont manufactured Type 558 x-ray film, which was more sensitive than the ORNL type, replaced the Type 552 packet on March 3, 1952. In the mid 1950s, Du Pont created the Type 555 film packet, which replaced the Type 558, and became the dosimeter predominately used at SRS. Beginning in March 1952, the Personnel Monitoring Section began to routinely calibrate beta-gamma personnel monitoring film. In January 1953, the Site could not yet process film badges. The dosimeters were worn for one week, collected, and transported to ORNL for processing. ORNL would then interpret the dose and forward the results to the Health Physics Group. The Site began to process film badges in March 1953 using the ORNL film badge dosimeter. The film had a minimum recordable dose of 30 mrem. The film was changed each week in the operating area and developed. The results were reported in mrem per week, manually recorded on individual exposure record cards, and kept in individual file folders. By early 1954, this operation required 45 people.

A new film badge dosimeter, designed by the Engineering Assistance Section, was introduced into service on November 9, 1959. This dosimeter contained a single packet of standard dosimeter film that could be inserted either manually or automatically using a specially made positioning jig. Manually processing film and data was a time-consuming practice, and human error was a concern because of the number of steps involved. To improve the efficiency of badge processing, an automatic dose computer was developed in 1959 by the Works Technical Department. This device was a combination densitometer and analog computer that determined film exposure. This computer performed satisfactorily in routine service for more than a year before it was replaced with a similar computer that used magnetic amplifiers. The new computer and reader accurately measured personnel exposures up to 1 R and 1 rad. Exposure data were recorded on IBM cards using a digital voltmeter and an IBM card punch. The reader was the key unit in the highly mechanized film badge handling/reading/recording system, which processed 3750 badges a week in the early 1960s.

In 1965, approximately 150 film badge dosimeters developed at SRS were modified to add LiF thermoluminescent dosimeters enriched to 99.91% Li-7, which was relatively insensitive to neutrons. This modification was made to evaluate the accuracy and reliability of TLDs if used for personnel monitoring. Based on previous studies, it was known that unshielded film overresponded twentyfold to 17 keV x-ray or gamma radiation compared to an over-response of 30% for the TLD. When placed in the film badge, the 30% over-response of the LiF was offset by a 0.15-inch plastic shield that effectively reduced the 17 keV radiation to 65% of its initial value. Film filtered with 1 g/cm^2 silver was known to overrespond twofold when exposed to 100 keV gamma radiation. Alternately, LiF dosimeters did not indicate any significant over-response for this energy.

On April 1, 1970, TLDs replaced film as the principal means of measuring external beta-gamma exposure. This change was based on the cost savings from extending low-exposure personnel to a quarterly badge cycle and the improvements in dosimetry accuracy from the near tissue equivalent response of the TLD regardless of photon energy. During this time, approximately 4600 employees were monitored by this system with 1600 participating in the quarterly cycle. This system was manually operated and used electronics and TLDs that were available commercially. Two ^{7}LiF chips were incorporated into the existing film badge. One TLD chip was positioned in the center of the open window, while the other was placed behind the aluminum shield (460 mg/cm^2) on the badge cover. This arrangement distinguished skin from penetrating exposures. These lithium chips were manufactured by Harshaw Chemical Company, and dosimeters were ordered in large uniform batches.

The TLD response to beta radiation is a function of energy, absorber thickness, and TLD self-absorption. The resulting skin dose depends on comparable factors where skin and outer clothing thickness absorb radiation incident on the body. An automatic, computer-controlled TLD badge processing system began operation in January 1973. Badge racks (25-badge capacity) that were compatible with the automated TLD system were used as pickup and deposit points for badges. They were located at convenient locations in areas where employees were assigned.

The TLDs were relatively insensitive to many of the environmental factors and chemicals to which film responded. The change from film to TLDs resulted in a significant reduction in exposure investigations. No significant differences in accumulated exposures were observed in any of the four general work areas during changeover from film to TLD dosimetry.

On July 1, 1982, the TLD badge was replaced by a commercial dosimeter manufactured by Panasonic. Frequent breakdowns of the one-of-a-kind badge reader used prior to this time made it difficult to process the number of TLDs that were in service (~4000 monthly and ~2500 quarterly). At times, personnel exposure data was not available for up to three weeks. The ability to meet impending DOE standards for dosimetry performance also was considered before adopting the Panasonic system. The new badge was one-third the size of the previous one.

Neutron Dosimetry (1951 to Present)

Monitoring personnel for exposure to neutrons began in 1951 using neutron track emulsion (NTA) film and the ORNL film badge. Initially, this film was replaced once per week and shipped to Oak Ridge for processing. On August 3, 1953, the Site assumed the responsibility of processing NTA film. The first badge used at the Site for personnel neutron monitoring was obtained from ORNL. It consisted of a film packet holder with an embossing plate for film identification, a 1-mm sterling silver filter, and a 1/2-inch diameter open window. The film packet holder was connected to a plastic badge equal in size to the SRS security pass. The dosimeter was exactly the same as the one used for beta-gamma personnel monitoring with the exception of the film. An Eastman Kodak NTA dental-size film packet was used in the neutron badge. The packet was inserted in the badge with the emulsion facing the back because it was thought that proton recoil yield from hydrogenous material would increase if the plastic faced the neutron source. A Site-designed film badge dosimeter replaced the ORNL dosimeter for personnel neutron monitoring in November 1959. Effective with the first exchange of NTA film badges after July 14, 1960, the badge cycle was extended to two weeks. The film was wrapped in aluminized mylar packets to prevent track fading from atmospheric conditions.

The procedure used for neutron dosimetry involved counting recoil proton tracks in NTA film. This method was accurate for neutron energies above 500 keV. The neutron film was managed separately from the beta-gamma film. Personnel responsible for reading the film (microscope observers) made all film changes and assisted in the darkroom. Upon return the film was then taken to the darkroom for developing. After developing, the film was read using a microscope to count proton recoil tracks produced in the NTA film emulsion. Considering only the shielded portion of the film, three sets of 40 fields were counted and the average number of tracks determined. By 1959 the NTA counting procedure had been modified. The tracks were counted on the calibration film; tracks produced in a blank film, an “error” figure (the statistical variation at 90% confidence level for the number of tracks observed) was subtracted for each location; and the number of tracks which corresponded to a 300 mrem exposure (“C” number) was determined. The dose recorded on personnel films was then determined by comparing the “C” number to the number of tracks on the film. Neutron film interpretation was never automated. During this time, badges were routinely interpreted by one technician. Approximately 400 NTA dosimeters were used per two-week badge cycle, and only 40-45 badges could be processed per day. Due to NTA film limitation in measuring low energy neutrons and the difficulties in interpreting this film when exposed to gamma fields greater than 0.5 R, an alternative neutron dosimeter had to be developed to detect lower energy neutrons produced by shielding effects in production facilities.

A prototype thermoluminescent neutron dosimeter (TLND), designed to detect albedo neutrons (mainly of thermal energy), was developed in 1968. Evaluation of the prototype for use in personnel monitoring was completed in October 1969.

The prototype TLND did not perform satisfactorily because of its large size and because the mounting clip did not maintain a consistent distance between the body and the dosimeter. This badge responded accurately (±20%) under laboratory conditions; however, it was found that a gap of 1/4-inch between the badge and the body could result in a difference as great as 48% in response. Thus, a new design was developed and completed in the spring of 1970.

A belt was incorporated into the design of the TLND to ensure a reproducible, consistent geometry during exposure in the field. This resulted in a snug fit when mounted against the wearer's body. The badge was modified further by reducing the badge size and weight by incorporating both pairs of TLD chips into to a single 2-inch diameter hemisphere. This arrangement covered the outer surface of the cadmium with stainless steel to increase the durability of the badge, providing a quick disconnect back plate to facilitate access to TLDs, and by modifying the thickness of the cadmium in the dome section to adjust the energy response of the dosimeter.

The redesigned TLND included a 2-inch polyethylene hemisphere that was sliced 1/4-inch from the rounded end (Hoy 1972). These two sections were separated by a cadmium shield plate. A pair of LiF chips, TLD-600 and TLD-700, was placed in the dome section of the hemisphere. Another pair of chips was placed in the hemisphere's base section. The curved surface of the polyethylene hemisphere was covered with a 1/32-inch-thick layer of cadmium, except for a 1/2-inch-radius area in the center of the dome, which was only 0.003 inch thick. This thin area allowed a small fraction of the incident thermal neutrons to reach the dosimeter chips in the smaller section of polyethylene. The cadmium was topped with a shell of 20-gauge stainless steel for protection. The unit was held together by a stainless steel back plate that has slots for attaching a belt to the TLND. When assembled, one pair of LiF chips was held in the dome section of the badge and was entirely surrounded by cadmium. The other pair was shielded by cadmium from the front only. The unit was designed with a small hole in the edge of the assembled badge to lock the unit together. A minimum recordable dose of 10 mrem was used for the TLND throughout its entire use period. Beginning January 1, 1995, the Hoy TLND was replaced with the Panasonic UD-809 albedo neutron dosimeter. The primary reason for the change was the difficulty involved in maintaining quality control for large groups of batch TLD chips, resulting from increased personnel neutron monitoring. This change permitted automated processing of personnel neutron dosimeters.

The UD-809 contains four elements shielded with cadmium and tin, three of ^{6}Li and one of ^{7}Li. The dosimeter is housed in a plastic case and covered with a plastic badge cover. It is worn in conjunction (i.e., issued in the same holder) with the Panasonic UD-812 beta-gamma dosimeter. Technical Specifications of this dosimeter are given in site developed technical manuals. While the Panasonic UD-809 is not as sensitive as the TLND and requires facility correction factors, it has the following advantages:

- Use of individual dosimeter calibration factors.
- Can be read in the same reader as beta-gamma dosimeters.
- Dosimeter results and their algorithms are processed via the same computer as the beta-gamma dosimeters.
- The dosimeters are not as bulky as the TLND.

As a result, the quality control and cost-effectiveness of the neutron dosimetry program has been improved, and many of the issue/control problems have been eliminated. The minimum reportable dose for the Panasonic UD-809 is 15 mrem.

Criticality Dosimetry

The Site has never experienced a criticality accident. However, because operations involve processing fissionable materials, the potential for a criticality accident to occur does exist. Although the chances of an accident of this type are very low because of various engineering and administrative controls, the radiation dose received by personnel must be quickly determined if an accident were to occur. A criticality dosimetry program has been in place since 1960 to assess an accident dose. Before 1960, special combinations of gamma- and neutron-sensitive film along with neutron-sensitive ionization chambers could have been used to measure gamma exposures from 20 mR to 1000 R and neutron exposures up to 1 rem. However, a criticality accident could have possibly resulted in radiation exposures greater than the upper sensitivity limits of these instruments. The Site developed the criticality neutron dosimeter (CND) in the late 1950s. A three-phase dosimetry system was established over the years to respond to a criticality accident. In the first phase, all potentially exposed personnel are screened. Indium foils in the personnel dosimeter and security badge would be activated by neutron exposure. The second phase involves approximating the neutron dose by analyzing ^{24}Na in blood. The third and final phase consists of a more accurate dose determination using a dosimeter capable of measuring dose over a wide range of energies. The CND's design has been modified over the years to improve its functionality.

The CND's components were assembled in a 3.5-4 inch long by 0.5-inch-diameter plastic tube. A clip is attached so it can be placed on the wearer's pocket. Indium, copper, and cadmium foils were shaped into hollow cylinders to lessen directional effects. These foils, along with specific amounts of sodium fluoride and sulfur, are contained in three small polystyrene vials. These materials are pre-weighed to expedite processing after an accident. The neutron fluence would be determined for five neutron energy levels based on the activation of indium, copper, and sulfur. Fluence values are then corrected for the direction of exposure because the CND is worn on the front side of the body, and activation is affected by body shielding and moderation. If left uncorrected, the dose estimate may be low by a factor of 2 for instances where the exposure is received from the backside of the body. The neutron fluence is corrected by comparing the amount of ^{24}Na in the blood of the exposed individual to the amount in the sodium fluoride in the dosimeter. The resulting value is compared to an experimentally derived ratio that provides the relationship between sodium activation in the blood and CND, neutron energy, and orientation of the dosimeter. The total neutron dose in rads is determined by multiplying the fluences in the five energy ranges, as determined from counting CND materials, by the respective dose conversion factors for that energy interval, then summing the five doses. The neutron dose is reported in rad since the quality factor for neutrons in a criticality accident has not been established. A lithium fluoride TLD, contained in a polyethylene vial, is used to measure gamma dose ranging from 25 mR to 10^6 R. Gamma dose measurements are corrected for direction of exposure.

Measurement Quality

Over the years, the Site has participated in many formal and informal intercomparison programs to assess the performance of its dosimetry program against other facilities. Site results usually were formalized in reports to management. In addition, an internal quality assurance and control program has been developed to ensure the accuracy of reported measurements.

A routine audit program was started in 1955 to verify the accuracy of the beta-gamma and neutron film badge monitoring programs. The audit program consisted of introducing a group of previously exposed badges (blind spikes) into the routine monitoring program. Originally, film exposed to known radiation levels was placed in visitor badges, which were then processed normally. Fictitious names were placed on badges to avoid entry of false records into the system.

As the dosimetry program evolved, the blind spike audit remained as an integral part and is still used today.

Calibration dosimeters have always been processed with film and TLD badges. These badges were interspersed with regular badges during processing to verify that the film or TLD reader was operating properly. This on-line quality control check is still used today.

Department of Energy Laboratory Accreditation Program (DOELAP)

In the early to mid 1980s, the ANSI Review Committee was charged with establishing performance standards for external dosimetry. Out of this effort evolved the National Voluntary Laboratory Accreditation Program (NVLAP). NVLAP participants were tested against ANSI standards and accredited in dosimetry services. Because of technical differences of opinion, DOE pursued developing its own performance standard for external dosimetry and accreditation program called DOELAP. Preliminary testing for DOELAP performance was conducted in the mid 1980s. The Site participated in these tests when DOELAP was finalized in the late 1980s. The Site was accredited and has maintained that accreditation, having received its most recent biennial accreditation in August 1999.

Presently, the Site participates in a Quarterly DOELAP/NVLAP External Dosimetry Processors TLD Badge Intercomparison Program. Two DOE facilities presently participate in the program, which serves as a site-specific blind spike check on TLD badge processing and irradiation protocols.

Endnotes

1. The historical information contained in this proceedings paper was derived from WSRC-RP-95-0234, *A History of Personnel Radiation Dosimetry at the Savannah River Site*.

Biography

Dante' W. Wells has been the Group Technical Lead for the Savannah River Site external dosimetry program since 1992. He was the Manager of the Internal and External dosimetry programs at Fernald in Ohio from 1988 to 1992. Prior to that he was the corporate radiation specialist for Entergy in Mississippi (1985-1988) and the REMS site coordinator at the Carolina Power & Light's HB Robinson Nuclear Station (1982-1985). Mr. Wells received his BS in biology/health physics degree from Virginia Polytechnic Institute and State University in 1982. He is a Lead Department of Energy Laboratory Accreditation Program (DOELAP) assessor for the DOE Headquarters EH-51 section through the Radiological and Environmental Sciences Laboratory DOELAP Performance Evaluation Program Administrator.

The Evolution of Internal Dosimetry Bioassay Methods at the Savannah River Site

George A. Taylor

Abstract

Throughout the 50-year history of the Savannah River Site (SRS), its employees have engaged in many activities that potentially could result in an accidental intake of radioactive material. For that reason, radiation workers have always been monitored for internal deposits of radioactive material. An overview of *in vitro* and *in vivo* monitoring methods are provided, as well as computational methods used to calculate radiation dose.

Introduction

The Savannah River Site (SRS) has monitored radiation workers for internal deposits of radioactive material since the Site's inception. This monitoring is part of a comprehensive internal dosimetry program that also includes workplace and personal contamination monitoring, air monitoring, bioassay, dose evaluation, administrative scheduling and reporting, and regulation and oversight. Program objectives are to detect and assess intakes (quantity of radioactive material that passes through the nares, the mouth, of the skin) of radioactive material in the workplace. This paper will concentrate on the progression of the bioassay and dose evaluation programs at SRS.

Two methods of quantifying how much radioactive material is in the body exist. In the first method, an inference is made as to how much radioactive material is in the body from how much is excreted in the urine or feces. This is referred to as *in-vitro* bioassay. In the second method, an inference is made as to how much radioactive material is in the body from the amount of photon radiation (a particle of electromagnetic energy) emitted from the body. This is referred to as *in-vivo* bioassay, which includes chest and whole body counting (WBC). The method of choice depends on the biokinetics of the radioactive material in the body; in particular, the fraction of the intake excreted through the urine and the intensity and energy of any photons emitted.

In-Vitro Bioassay

Prior to completion of whole body counting facility in 1960, only *in-vitro* bioassay, primarily urinalysis (analysis of a urine sample), was performed. *In-vitro* urine bioassay for nuclear by-products performed during this period was typically a gross beta activity analysis. Since that time *in-vitro* bioassay analysis has become more radionuclide specific, and may be subdivided into the following categories:

- plutonium urinalysis
- tritium urinalysis
- uranium urinalysis
- trivalent actinide urinalysis
- neptunium urinalysis
- fission product urinalysis
- fecal bioassay

Plutonium Urinalysis

The first procedure for plutonium urinalysis (Sanders 1956) was implemented in 1954 and used until 1959. This procedure used bismuth phosphate and lanthanium fluoride coprecipitations to separate plutonium from 1500 mL of urine. The precipitate was resuspended in acid and electroplated on stainless steel disks. These disks were placed for one week on glass plates coated with Kodak film emulsions. The plates were developed and the alpha tracks were counted to quantify the plutonium. This analysis for plutonium had a minimum detectable amount (MDA) on the order of 0.035 disintegrations per minute (dpm) per liter of urine, which is comparable with detection capabilities available today.

The coprecipitation procedure was labor intensive. In 1959 (Sanders 1961), an ion-exchange technique was adopted that cut processing time in half and reduced the amount of urine used to 250 mL, while keeping the MDA about the same. This procedure, which also employed electrode position and alpha track counting, was used until about 1966. A reporting level of 0.05 dpm per 1.5 liters of urine was used for plutonium urinalysis reported during the 1954-1966 period (i.e., the "less-than" level was 0.05 dpm per 1.5 liters). Results were expressed per 1.5 liters of urine because Standard Man was assumed to excrete 1.5 liters of urine per day, which permitted (in theory) a direct conversion from concentration to excretion per day. Standard Man represented a set of agreed-upon values for the many characteristics of man that are needed for internal dose calculation.

In the mid 1960s, two significant changes were made to the plutonium urinalysis. First, in 1964, solid-state surface barrier alpha detectors (Butler 1966) were introduced to replace Kodak film emulsions. These gross-alpha detectors reduced the time for counting a sample from one week to one day and greatly simplified the counting procedure. Second, around 1966, the ion-exchange method was replaced with a liquid ion-exchange method, utilizing the organic extractant trifluoro thionylacetone or TIOA (Butler 1968, Butler 1965). The reporting level for the TIOA gross-alpha method was 0.1 dpm per 1.5 liters of urine. With few changes, this method was used for plutonium urinalysis until 1988, a remarkable span of over 20 years.

In the early 1980s, experimentation began with a coprecipitation technique developed by Kressin (1981). This method used alpha spectroscopy, which permits the addition of Pu-242 tracer into a sample to determine radiochemical recovery of plutonium and detector counting efficiency. With gross alpha counting, only approximate "batch" estimates are made of these parameters. Up until about 1988, only routine urine samples were analyzed by the alpha spectrometry method and the TIOA ion exchange method was used for special urine samples. Reporting levels for alpha spectrometry during this period were 0.05 dpm per 1.5 liters for Pu-238 and 0.07 dpm per 1.5 liters for Pu-239. Around 1988, the coprecipitation step of Kressin's procedure was dropped to give the procedure that is in use today.

Since startup, *in-vitro* bioassay results were recorded on cards that were placed in the individual's personnel file. Starting in 1990, bioassay results were stored on a computer. At the same time, all plutonium results were converted from dpm per 1.5 liters to dpm per liter.

Tritium Urinalysis

From startup to 1958, tritium oxide in urine was analyzed by adding calcium metal to the urine and running the evolved hydrogen through an ionization chamber (Hursh 1958). The MDA for this method was 1 µCi/l. In 1958, an automatic sample changing liquid scintillation counter (LSC) was procured (Hursh 1958). This counter was much easier to use and, with a one-minute count, provided the same MDA as the ionization chamber method.

Through the years, the detection capability of the LSC method improved, but the 1 µCi per liter was retained as a reporting level. The reporting level was lowered, first to 0.5 µCi per liter and later to 0.1 µCi per liter, which is the level in use today.

Uranium Urinalysis

Analyzing uranium by measuring alpha radiation emitted is referred to as an analysis for "enriched uranium". The first method used for enriched uranium involved coprecipitation followed by electrodeposition (depositing a substance at an electrode using a direct current) and Kodak film emulsion counting of alpha radiation (Boni 1960). This method had a reported sensitivity of 0.15 dpm per 1.5 liters of urine. In the mid 1960s, the TIOA/gross alpha counting method was adopted for enriched uranium analyses (Butler 1968). This method had an MDA of about 1 dpm per 1.5 liters, which was considered adequate at the time.

The analysis of uranium based on its chemical properties is referred to as a "depleted uranium" analysis. Analyses for depleted uranium were performed with the Oak Ridge fluorophotometric method from 1954 to 1982. This method involved using a spectroscope to measure total uranium.

The delayed neutron analysis (DNA) method (Hurley 1982) was adopted for both enriched and depleted uranium analyses around 1982. This method involved coprecipitating the uranium with calcium fluoride, activating the sample in a reactor, and counting the delayed neutrons emitted by the U-235. This procedure had an MDA of 0.14 ng of U-235, which provided a 1 µg per liter MDA for natural uranium and a 1 dpm per liter MDA for enrichments typically encountered at SRS.

With the shutdown in 1986 of the reactor facility used for DNA of uranium, the TIOA method was again adopted for enriched uranium and the Jarrell-Ash method (x-ray fluoroscopy) for depleted uranium. In 1990, an ion-exchange method was adopted for enriched uranium. This method, which is still in use today, uses alpha spectrometry with an internal U-232 tracer.

Kinetic phosphorimetry analysis (KPA) for depleted uranium was used from 1986 through 1994. Since 1994, both enriched and depleted uranium have been analyzed by the ion exchange/alpha spectrometry method.

Trivalent Actinides

The trivalent actinides consist of americium, curium, and californium. There was increased production of these materials at SRS in the early 1970s. Analysis was accomplished by extracting the trivalent actinides from the plutonium-TIOA raffinate with di,2-ethylhexylophosphoric acid (HDEHP) followed by gross alpha counting. Later, the HDEHP extraction was replaced with a organic extraction that was less complex. The procedure required 500 ml of urine and had an MDA of 0.3 dpm per 1.5 liters.

In 1990, ion exchange resin was used to separate the trivalent actinides from other actinides. The sample was direct mounted (placed on a planchet with rimmed edges and heated to evaporation), and was "gross-alpha" counted by using a wide window on the alpha spectrometer. This method can achieve MDAs on the order of 0.1 dpm per liter. This is a gross-alpha technique and therefore does not use an internal tracer.

Since 1994, extraction chromatography resin has been used to separate the trivalent actinides from other actinides. With the implementation of improved software and use of an Am-243 tracer, chemical recovery and counting efficiency may be determined for each sample.

Neptunium Urinalysis

Starting in 1959, neptunium was coprecipitated, ion exchanged, electrodeposited, and counted on Kodak film emulsion. In the mid 1960s (Butler 1968), the TIOA/gross alpha method was adopted. This method was replaced in 1993 by anion (negatively changed ions) exchange followed by direct mounting and gross-alpha counting.

Since 1994, extraction chromatography resin has been used to separate neptunium from fission products and other actinides, and electrodeposition has been used to mount the sample. There are no suitable isotopes of neptunium available to use as tracers, so this is still a gross-alpha counting technique.

Fission Product Urinalysis

Strontium urinalysis was performed in the 1950s and 1960s by coprecipitation followed by beta counting on a Geiger Mueller counter. Since 1969, strontium has been analyzed by a liquid ion-exchange method that extracts the Y-90, which is counted on a beta proportional counter. In 1998, the strontium method was modified by using an ion exchange column to extract Sr-90, which is also counted on proportional counter. Gamma-emitting fission products have historically been analyzed by gamma-spectrometry with sodium iodide detectors.

Fecal Bioassay

The history of fecal analysis is somewhat fragmented. It appears that a modified TIOA procedure (DPSOL 1972) was used in the 1970s for fecal analysis of actinides, but it is uncertain when the procedure was adopted or abandoned. The primary method for fecal bioassay used in the early and mid 1980s was gamma-spectrometry with a phoswhich detector. Some of the samples analyzed by this method were subsequently dried and sent to offsite laboratories for analysis. Since 1994, fecal samples have been analyzed by a modified urinalysis procedure.

In-Vivo Bioassay

The history of *in-vivo* bioassay at SRS may be broken down into three periods that are separated by the introduction of new detector technologies. The first period of *in-vivo* bioassay began with the development of large solid scintillation detectors made of sodium iodide in the 1950s that were used to measure high-energy photon emitters in the body. The development of low-energy phoswich detectors, which made detection of low-energy photon emitters feasible, began the second period in 1970. Finally, the modern period began at SRS in the late 1980s with the introduction of high-resolution germanium detectors.

Whole Body Counting

In-vitro bioassay was the only method available to assess occupational intakes of radionuclides in defense facilities before the mid 1950s. During this period, *in-vivo* bioassay became feasible with the introduction of large sodium iodide detectors at Argonne National Laboratory and 4p liquid scintillator detectors, which made it possible for radioactive emissions emerging from the top and bottom surfaces of a sample to be counted, at Los Alamos Scientific Laboratory. Before the development of these detectors, *in-vivo* bioassay was performed with ionization chambers or Geiger-Muller tubes that were insensitive and not practical for assessing occupational intakes of radionuclides. In 1957, Sanders (1965) proposed that a whole body counting facility similar to that at Argonne utilizing sodium iodide detectors be built at SRS.

This state-of-the-art whole body counting facility was completed in 1960. The room, known as the steel room, was built of 12-inch-thick pre-WWII steel that was originally fabricated as armor for a battleship. Pre-WWII steel was selected because it did not contain man-made radionuclides (fallout Cs-137 and Co-60 from smelters) that can increase the background in the room. The steel room is 8-1/2 feet wide by 11 feet long by 8 feet high, weighs 155 tons, and is lined with 1/8-inch low-radioactivity lead sheet.

The first detector used in the steel room was an 8-inch diameter by 4-inch-high cylindrical sodium iodide detector. The detector was covered with 1/2-inch of lead (except for the bottom) and had three photo multiplier tubes, devices used to convert weak light output of a scintillation pulse into a corresponding electrical signal. This detector had an effective energy range of 100 keV to 2000 keV, which meant that it could not be used to detect or quantify radionuclides like plutonium and americium that emit only low energy (<100 keV) photons.

The counting geometry, or source and detector position, used was the 40-cm arc. It was also known as the Argonne chair. The person sat in a reclining chair and the detector was suspended above the pelvic area. The chair geometry placed the body from chin to knees approximately 40 cm from the detector. Limited investigational counts of the lungs were available with the detector placed in contact with the chest.

The count data was collected in a 200-channel multichannel analyzer. Analysis of the count data with an IBM 1620 computer (Watts 1963) began in 1963. The 200 channels were divided into 15 energy regions that represented photons of various energies. The background in these regions was determined for a number of individuals who had only varying amounts of natural K-40 and Cs-137 fallout in their bodies. From these data, a "prediction equation" was calculated that was used to estimate the background in the corresponding energy regions for a worker whose body may have contained other radionuclides.

The whole body counter was calibrated with the REMCAL phantom, a butyrate plastic phantom (geometric model) designed to visually resemble man. An accepted practice at this time was to calibrate whole body counters with volunteers who were administered small quantities of radionuclides. This technique was never used at SRS, but intercomparisons were done with other defense facilities that did. SRS was within 2-40% of their calibrations for radionuclides like Cs-137.

The MDA was defined to be a count that exceeded three times the standard deviation (3s) of the expected count rate in an energy region. The MDAs calculated for various radionuclides during this time are shown in Table 1 along with the corresponding maximum permissible body burdens. As indicated in Table 1, even the very first whole body counter was capable of detecting small intakes of high-energy photon emitting radionuclides.

Sometime during the early 1970s, the chair geometry was replaced with a bed geometry. Shortly thereafter, the single 8-inch by 4-inch sodium iodide detector was replaced with five 4-inch by 4-inch cylindrical sodium iodide detectors placed under the bed. The sodium iodide detectors were positioned in an arc under the bed to provide constant efficiency for all detectors. In essence, the detectors rather than the person were placed in an arc. Count data continued to be analyzed by the 200-

Table 1. Minimum Detectable Levels for SRS *In-Vivo* Counting Whole Body Count with 4 x 8 Crystal

Isotope	Minimum Detectable Level (20 min count) (nCi)	1% MPBB Values (nCi)
^{144}Ce	29	50
^{Nat}U	62	.05
^{131}I	1.4	7
^{106}Ru	6.1	30
^{137}Cs	1.0	300
^{95}Zr-^{95}Nb	2.2	200
^{65}Zn	5.1	600
^{140}Ba-^{140}La	9.3	40

All 4 x 8 crystal "whole body" count MDAs are based on +3 deviation from average clean person channel MCA.

In the mid 1980s, commercially available standup counters using large sodium iodide detectors and a shallow shield arrangement (FASTSCAN) were obtained to provide 2-to-4-minute counts for high-energy photon emitters. Two of the counters were mounted in trucks so that they could be moved from place to place as needed.

Chest Counting

The whole body counter, useful for monitoring workers for intake of gamma-emitting fission and activation products, was of no use to detect or quantify intakes of many actinides such as plutonium and americium. Efforts had begun at SRS in the mid 1960s to develop low-energy photon detectors that would detect actinides in the chest. Most efforts involved either thin sodium iodide detectors or xenon-filled proportional counters. The xenon proportional counters, with their high resolution, showed great promise but were eventually abandoned because of their high background and low efficiency.

Development of the thin sodium iodide detector continued. In 1966, a 1-mm thick by 5-inch diameter detector with a 0.001-inch window was first used to count workers involved in a contamination incident. Thin sodium iodide detectors were used with varying degrees of success from 1966 until 1971, when they were replaced with phoswich "phosphor sandwich" detectors, composed of a layer of sodium iodide on top of a layer of cesium iodide.

This was a major breakthrough in *in-vivo* bioassay for low-energy photon. This detector's thin construction and inherent anti-coincidence counting (the elimination of undesired radiation) capabilities greatly reduced the background in the low-energy region of the energy spectrum. In 1972, a dual phoswich system composed of two 5-inch detectors was placed in service. Although improvements were made in data analysis and calibration over the years, this basic system was used for chest counting for nearly 15 years until the adoption of planar germanium detectors in the late 1980s.

The detection capability of the phoswich detectors for plutonium is somewhat uncertain. In 1969, researchers at Los Alamos Scientific Laboratory (Dean 1969) claimed an MDA of approximately 1 nCi for Pu-238. During the 1970s, measurable Pu-238 chest burdens on the order of 4 to 10 nCi were reported at SRS. Currently, however, estimates of the MDA for Pu-238 using state-of-the-art germanium detectors is approximately 60 to 70 nCi. The cause of this discrepancy is not known, but it may be related to differences in the way the MDAs were calculated in the 1970s versus today.

In the late 1970s, small coaxial germanium detectors were installed in the steel room. These detectors were pointed at the sides of the chest to detect the 186 keV photon from the U-235 in enriched uranium, which was not readily detected by the phoswich or sodium iodide detectors. These detectors were also used to identify high-energy photon emitters, taking advantage of the superior resolution of the germanium detectors. In the mid 1980s, thick phoswich detectors specifically designed for enriched uranium detection were purchased. This turnkey system was only used for a few years before the germanium chest counter replaced it.

Routine use of a six-detector germanium chest counter began at SRS in December 1989, replacing the phoswich system. The detectors were 2000 mm^2 surface area, housed separately in "organ-pipe" dewars (Canberra ACT-I design). A reclining chair counting geometry was used. In August 1995, a pressure transient damaged the detector array. Because of this, a planned move to a new facility was accelerated, and new 2800 mm^2 detectors, two each housed in a 7-liter dewar (Canberra ACT-II design) were placed into service in September 1995.

Chest Wall Thickness

The thickness of the chest wall is an important factor in quantifying actinides in the chest. In 1970, ultrasound equipment was purchased to measure the chest-wall thickness. Through the years, the equipment was abandoned and upgraded several times. Eventually, an empirical height-weight to chest wall thickness algorithm was adopted to estimate the thickness of the chest wall. This algorithm, which is still in routine use today, is known to be inaccurate for females. Presently, in cases which intake evaluation is based on chest count bioassay data, direct measurement of chest wall thickness can be made by ultrasound techniques.

Calibration

To this day, a major problem with chest counting is the accurate calibration of the system. The early thin NaI detectors were calibrated with simple sources consisting of a suitable radioactive material plated on metal planchets. In 1969, a human skeleton with tissue-equivalent lungs was first used for calibrations. This phantom was used until the Livermore phantom was adopted in 1982. The Livermore phantom was designed and prototyped by *in-vivo* experts in the weapons complex. After a series of round-robin intercomparisons in the complex with the phantom in the late 1970s, the phantom was turned over to commercial firm for mass production. Up to that time, all sites in the complex were using different phantoms of their own design and construction. Thus, with the introduction of the Livermore phantom, for the first time a standard chest phantom was readily available to anyone with enough money to buy it (~$20,000). Over the years, this *de facto* standard has greatly improved the consistency of chest counting throughout the complex. More significantly, the success of the Livermore phantom in the early 1980s started a move at SRS and in the complex to "turnkey" commercial phantoms, detectors, and software that have essentially eliminated in-house research and development in the area of *in-vivo* bioassay.

Internal Dose Computational Methods

Assessment of internal dose for workers in the nuclear weapons complex has always been based on the evaluation of bioassay data rather than air monitoring data. The methods used to evaluate bioassay data throughout the history of SRS have changed to keep pace with improvements in the technology of internal dose assessment and the evolution of internal dose regulations. All radionuclides have not been impacted equally as things have changed. For example, the methods used to evaluate intakes of tritiated water have not changed significantly in over 40 years whereas the methods used to evaluate intakes of plutonium have changed significantly.

Calculating Body Burdens

From plant startup until 1975, internal dose limits were expressed in terms of the MPBB or maximum permissible organ burden (MPOB). The body or organ burden for radionuclides that emit high-energy photon radiation and target specific locations in the body, like Cs-137 and I-131, are directly determined by *in-vivo* bioassay. No calculations are required. No significant intakes of these materials has ever occurred at SRS, with few measured burdens exceeding a percentage of the MPBB.

Intakes of radioactive materials that cannot be quantified by *in-vivo* bioassay are more difficult to evaluate because inference must be made to what is in the body from what is coming out. The standard practice for evaluating intakes of these materials (DPSOL 1968 and DPSOL 1968a) except for tritiated water during the 1954-1975 period was to:

- collect enough excretion data, usually urine, to define the excretion curve
- integrate the excretion curve from the time of uptake to infinity to calculate the total amount of material excreted through the urine

- adjust the integral for the fraction of an uptake excreted by pathways other than urine to determine the uptake

The excretion data were typically fit to simple power functions of time like

$$A = Et^{-b},$$

where A is the amount of material in the urine at time t, and E is the amount of material in the urine at t=1. E and b were determined in the least squares fit. The power function was popular at this time because it can describe multicomponent excretion curves and be readily transformed into a simple linear form that is easy to manipulate mathematically without the aid of sophisticated computers. If few data were available, default values were used for the exponent b such as -0.74 for plutonium, and the available data were used to determine E. Evaluations from this period that appear in personnel files are typically not well documented as to the exact methods used. However, it can be safely assumed that the power function method was used because no evidence supporting the use of another method has ever been found.

Notice that the power function method gives an uptake, which is the systemic body burden shortly after the time of the intake. Material in the lungs or in a wound was not accounted for and was usually ignored until it eventually became systemic (absorbed in the blood). Starting with the installation of the dual phoswich chest counter in 1971, it became possible to directly measure chest burdens as long as they were quite large. Measured chest burdens were compared with the maximum permissible lung burden (MPLB) to determine their significance.

Whole Body Dose from Tritiated Water

Tritiated water is unique among internal emitters at SRS because intakes of it have always been evaluated in terms of whole body dose equivalent that has been added to whole body dose equivalent from external sources. This was true even during the 1954-1975 period when limits were expressed in terms of the MPBB. The basic technique for calculating dose from tritiated water urinary excretion data has changed little over the years. For an acute intake of tritiated water it consists of

- fitting the urinary tritium concentration data to a single exponential
- integrating the urinary concentration curve
- multiplying by a constant to get dose.

The primary differences between the current method, which is based on Publication 30 of the International Commission on Radiological Protection (ICRP 30), and earlier methods, which were based on ICRP 2, are the

- default biological half-life (12 days then, 10 days now)
- target tissue (43 kg of body water then, 63 kg of soft tissue now)
- default mass of body water (43 kg then, 42 kg now)
- quality factor for tritium (1.7 then, 1.0 now)
- mean energy of tritium beta particle (6.1 keV then, 5.7 keV now).

Note that the default half-life is used only to interpolate between samples under a chronic, or recurrent, intake pattern. For acute intakes, the person's biological half-life, the time at which one half of a given substance is eliminated by biological processes was determined from his excretion curve. The ICRP 30 methodology for calculating tritium dose, especially the quality factor and target tissue, was adopted in 1986 (Reinig 1986). Changes in the default biological half-life and mean beta energy were probably made before 1981.

The exact methods used to calculate dose from chronic exposure to tritiated water prior to 1980 are not known for certain. In the days prior to computer evaluation of the data, only results greater than 5 μCi/l were evaluated (Reinig 1963), probably by a linear interpolation method (La Bone 1992). As computers became available, all results were evaluated in terms of dose by an exponential interpolation method (Boone 1994).

Organ Doses

From 1975 to 1987, limits for internal emitters were given in terms of organ doses in a year. Organ dose equivalent (H_T) over the time period from t_1 to t_2 was calculated using the following formula (Hall 1980):

$$H_T = \frac{1.87 \times 10^4 \, q \, \varepsilon \, f}{m} \int_{t_1}^{t_2} e^{-\lambda t} \, dt,$$

where

q = body burden at time t_1,
f = fraction of body burden in organ, and
e = effective energy for radionuclide in organ.

Although this is an ICRP 2 type calculation, the referenced document, which was issued to document the first Annual Exposure Reports, uses many ICRP 30 parameters. For example, the f for plutonium in bone is 0.45, which is the parameter given in ICRP 30. As new internal dosimetry methods evolve, it is quite common for parts of the new methods to be incorporated into the old methods. This is as true today as it was in 1960 and is a natural consequence of trying to use the most current technology.

Intake Assessment

As discussed earlier, ICRP 30 methods began to be incorporated into existing intake evaluation procedures in the late 1970s and early 1980s. In ICRP 30, the concept of calculating an intake from bioassay data and using this intake to calculate a 50-year committed effective dose equivalent (CEDE) was introduced. In 1987, the contamination cases of all active employees who had intakes of radioactive materials other than tritium were re-evaluated using ICRP 30 methods. All of these individuals were informed of the reassessment and were given the new dose estimates. DOE Order 5480.11, which required the calculation of annual and CEDE, was adopted in 1989. *The DOE Radiological Control Manual,* which required the exclusive use of CEDE, was adopted in 1993.

Work continued on the reevaluation of the cases of all the non-active (retired, terminated, or deceased) employees, and work was completed in 1992. These individuals were not informed of the reassessment unless they requested dosimetry information after 1987. Thus, as of 1992, all known intakes of radioactive materials other than tritium had been reassessed with ICRP 30 methods.

The methods used to evaluate bioassay data to calculate intakes and dose are documented in the *SRS Internal Dosimetry Technical Basis Manual*. The methods used to evaluate bioassay data have not changed since 1987. Internal doses from radionuclides other than tritium will be continually refined as new and improved biokinetic and dosimetric models are introduced and additional bioassay data are collected. Because intakes of tritiated water have always been addressed in terms of whole body dose, historic intakes of tritiated water are not reevaluated to incorporate the newest models. This is the same policy used for historic external doses, which are not reassessed.

Chelation Therapy

Once an intake of a transuranic (plutonium, americium, and curium) occurs, there are only three therapeutic procedures available to reduce the intake and mitigate the dose: excision of material from wounds, removal of the material from the lungs with lavage, and chelation therapy. Chelation therapy enhances the urinary excretion of transuranics from the body by binding of a chelation agent to transuranic elements in the bloodstream. Hall et al. (Hall 1978) published a mathematical model for evaluating intakes of plutonium after chelation therapy with diethylene-triamine-pentaacetic acid, better known as DTPA. This model was derived from bioassay data of workers at SRS who had received DTPA treatments. Prior to the publication of this model, intake evaluation following chelation therapy was not possible using the conventional methods.

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Biography

George A. Taylor began his professional career at the Savannah River Site in 1989 after receiving a MS in Health Physics at Texas A&M University. From 1989 to 1991 he was assigned to Environmental Monitoring where he was responsible for issuing various environmental reports to DOE and State officials. He also authored the Site's annual Environmental Monitoring Report and managed the environmental TLD program. In 1992 he became a member of the Field Technical Support group in Health Physics Technology. He has developed several site-wide technical guidance documents that have been incorporated into the SRS radiological control program. In 1995 he captured historical dosimetry practices in "A History of Personnel Radiation Dosimetry at the Savannah River Site". Since joining Health Physics Technology, he also has held technical support roles in Internal Dosimetry Programs and Instrument Programs.

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Environmental Radioactivity On and Near the Savannah River Site Before the Start of Nuclear Operations

William C. Reinig

Abstract

In the spring of 1951, a few months after the start of construction of the Savannah River Plant and now Savannah River Site, a small team of scientists and engineers began a survey of environmental radioactivity at and near the Site. Never before had such a study been conducted prior to the startup of a nuclear facility. The 18-month survey covered 6000 square miles in South Carolina and Georgia. A principal objective was to characterize environmental radiation and radioactivity so that any increase after the beginning of nuclear operations could be readily determined. The survey was to be the prototype for the environmental monitoring program that was to continue after nuclear operations began. Air, surface and subsurface water, vegetation, soil, and other components of the environment were analyzed for radioactivity. Fallout from nuclear weapons tests complicated the study. The survey met its objectives and inaugurated the long tradition of environmental stewardship that has served the Site and its neighbors so well.

In October 1950, the Du Pont Company accepted President Truman's request to build and operate the Savannah River Plant (SRP) for the Atomic Energy Commission (AEC). Du Pont learned from operating a government-owned nuclear plant during World War II that a comprehensive environmental survey before its startup would have been a valuable asset. In the spring of 1951, Du Pont began planning a preoperational survey of natural radioactivity on and near SRP. The study had several purposes. One was to characterize the environmental radiation and radioactivity so that any increase over this baseline could be readily determined and controlled as necessary. Another purpose was to serve as a prototype for the long-term monitoring program that was to follow the preoperational survey.

This was a trail-blazing study. Never before had such a survey been carried out prior to the startup of a nuclear facility. No federal or state regulations mandated it. Years later these surveys became a requirement for nuclear plants in the United States and in many other nations. It is difficult to realize that in the 1950s the thousands of federal and state environmental regulations now on the books didn't exist then. The establishment of the Environmental Protection Agency was still 20 years away. In those earlier days when you said "ecology" you had to explain its meaning.

In the spring of 1951, C.M. Patterson, who had been a leader of Du Pont's radiation protection program at the Hanford Plant during World War II and had recently rejoined the Company, began recruiting a team of scientists and engineers to conduct the SRP environmental survey. The pool of experienced candidates was limited. Only a few AEC contractors were monitoring radioactivity in the environment, and their programs were relatively small. Throughout the nation, probably fewer than 30 scientists and engineers were involved in these activities. Several graduate programs in health physics sponsored by the Atomic Energy Commission had begun a year or so earlier. The team, designated as the Site Survey Group, that assembled at SRP during the summer of 1951 was a mix of people from other AEC sites, universities, and Du Pont plants. I was the team leader.

A year later, C.M. Patterson moved to SRP from Du Pont headquarters in Delaware and was appointed the first head of the Health Physics Section. The Site Survey Group became part of his organization. He directed the successful radiation protection program at the Site until his retirement in 1978.

Ralph Gosline (from Los Alamos) and I (from Brookhaven National Laboratory) were the first team members to arrive at SRP. When we came in June, road construction and the clearing and grading efforts for building sites had just started. Most people still remained on their farms and in their homes in the towns of Ellenton and Dunbarton and several other smaller communities on the Site. Some construction workers lived in tents near these towns because lodgings were scarce and expensive. Stores and other businesses were open. Farmers were growing cotton, corn, and peanuts. Trains still stopped at the Ellenton station. Within a year, the 1500 families who resided in the 300-square-mile area would be gone, with many moving their houses with them. When the environmental survey was completed in 1953, only traces remained of the towns and farms where 6000 people once lived.

About a dozen Du Pont engineers and chemists, who were to work in a pilot plant beside the river, were already here when we came. They were following the plant's construction, scheduled for completion in the fall of 1951. Several laboratories in the construction effort were assigned to the Site Survey Group.

Upon arrival, one of our first tasks was to learn about the Site. It was a difficult area to get to know. Although we had excellent maps from the Army Map Service, they didn't show the impenetrability of marshes and swamps that bordered practically all of the 22-mile stretch of Savannah River that adjoined SRP. The maps didn't indicate the thick briar undergrowth and tall canes that made access difficult to the 75 miles of streams on the Site. The maps didn't show that many of the unpaved roads had clayey surfaces that became slick when wet. Neither did the maps indicate that most roads would soon be clogged with construction vehicles and houses being moved off the Site. Of course, they didn't warn about the alligators and poisonous snakes. We couldn't fully appreciate from the maps the isolation of the river with hardly any traffic and only a few landings on either the Georgia or South Carolina sides for almost a hundred miles downriver.

On July 26, 1951, Ralph Gosline dipped a bottle into the Savannah River at Gray's Landing. A day or so later the water was analyzed for radioactivity in a small windowless construction shed. This was the start of the preoperational study of environmental radioactivity. It was also the birth of the Site environmental monitoring program that has continued for a half century.

As the summer progressed, new members of the monitoring team arrived. At the end of the summer, the team had 12 members. It was the first operations group on the Site, although about 2000 construction employees had already arrived. A few years later the construction force grew to 40,000 workers.

Since the preoperational survey was to be a dry run for the routine monitoring program after SRP startup, its organization and content reflected our concepts of the post-operational monitoring. We anticipated that the reactors and the separations facilities would be the primary potential sources of environmental radioactivity. These facilities were to be built near the center of the Site to provide a buffer zone of about 10 miles between the facilities and the Site boundary. Radioactive releases would first be monitored in facility stacks and pipelines and by monitoring stations immediately outside the buildings. We placed additional monitoring stations in a ring around the area containing the reactors and separations plants and in another ring farther away at the Site boundary. Finally, there were stations 25 miles from the Site. These stations were either small buildings with devices to collect air and rainwater samples or places where soil and vegetation were routinely obtained. Each site stream was sampled at several locations, and the river was sampled at 16 points. At the stream and river locations where water was collected, sediment samples were also obtained. The density of the monitoring stations was greatest near the nuclear facilities and decreased with distance from them.

Crops from local farms and water supplies in cities and towns as far away as Savannah were also analyzed for radioactivity. We inventoried the many open farm wells and selected those that would be routinely sampled. These were preserved and barricaded. The remaining wells were filled in because they were a safety hazard for construction workers and the Site Survey team.

We anticipated that in the late fall atmospheric tests of American nuclear weapons might deposit considerable radioactivity in the area. Therefore, we pushed hard to collect appropriate samples and to install fallout-monitoring equipment during the summer and fall of 1951. Debris from unexpected Russian nuclear tests blanketed the area in October, several weeks after the air monitoring stations were put into service, two on the Site and one in Aiken. American fallout arrived in November. In addition to the air monitoring stations, large sheets of flypaper were placed at many locations to detect fallout. This low-tech, inexpensive method worked well. Fallout particles adhered to the paper. When the paper was placed on photographic film, dark spots on the film caused by the radiation from the particles were counted to get a relative measure of the fallout.

Before the completion of our laboratories, we processed samples in a construction shed that wasn't air-conditioned. We often worked outside during the summer of 1951 using nearby stumps of large trees as laboratory tables. We quickly learned that some instruments that performed well in the dry climates of Hanford and Los Alamos could not tolerate the high humidity at SRP. Until they were later moved into an air-conditioned laboratory, the instruments functioned only if we loaded them with a drying agent, which had to be replaced every morning. Not having hoods and other common laboratory services and frequent power interruptions beset the analytical program.

During the summer, we met other environmental organizations that were beginning their work at SRP. DuPont engaged the Philadelphia Academy of Natural Sciences to baseline the health of the river. Dr. Ruth Patrick headed the Academy's team. They rented several rooms in a motel in Allendale, South Carolina, and converted one into a laboratory. Another team, under the direction of Dr. Eugene Odum from the University of Georgia, was starting long-term terrestrial studies. His group was housed in a barn-like structure on the edge of the Site. These veterans of southern field studies scheduled their outside work between 4 p.m. and dark when it was cooler. The studies started by Dr. Patrick and Dr. Odum continue today. Dr. Patrick's work represents the longest continuous set of biological studies in an aquatic environment in the United States, and probably in the world. The studies of Dr. Odum's group expanded further, and they became the basis for establishing the Savannah River Ecology Laboratory in 1962.

In 1951, the Savannah River Advisory Board was established by the U.S. Surgeon General to monitor the effects on the river of the Savannah River Site and the Clarks Hill Dam, which was being built. The panel, representing federal and state agencies having jurisdiction over water resources, endorsed the concept of the preoperational survey. The Board routinely reviewed the plans and results of the study. This gave us an opportunity to discuss the Site's programs to protect the environment.

In those years, the media and the public were, of course, very interested in the Site. Du Pont had no public relations organization, and we were often asked by the local office of the Atomic Energy Commission to explain the survey and the Site's plans to safeguard public health and the environment. These outreach activities demonstrated to a wide audience the strong resolve of SRP to be a safe neighbor.

In January 1953, the 18-month survey was completed. Approximately 6600 environmental samples were analyzed. Radiation and radioactivity were characterized on and around the Site. Monitoring stations, equipment, instruments, procedures, and trained people for the post-operational environmental monitoring were in place. This allowed a seamless transition to the continuing monitoring program. When the final major facility came on line in 1955, the Site's routine monitoring program had been in operation for 30 months.

I will not discuss the results of the analyses of the environmental samples that were collected and analyzed for total radioactivity or for specific radionuclides. These are discussed elsewhere (Reinig et al. 1953). If the study had been made 20 years later, we would have determined the specific radionuclides in many more of the environmental samples using rapid and accurate alpha and gamma spectrometry. But this was not practical using the state-of-the-art methods of the early 1950s. The results of the analyses were generally about what we expected.

But there were surprises. For example, we found that the granite aggregate about to be used to construct laboratories for measuring low levels of radioactivity contained high concentrations of natural radioactivity. It was replaced with an aggregate that had a 100 times less radioactivity. Another surprise was the substantial amount of fallout from Soviet nuclear weapons testing that arrived at the Site in October 1951. While we anticipated the possibility of considerable fallout from American tests, we didn't expect much from the Soviet test half way around the world.

One of the purposes of this prototypical study was to uncover and solve problems so that they would not be encountered later (Patterson 1987). Experience with boats on streams and the river indicated special emphasis on boat safety was needed. This training prepared the crews to respond safely to unexpected events, such as when a snake dropped into a boat from an overhanging branch or when a motor failed far downriver beyond the range of their radio. We discovered that birds damaged the instruments that measured environmental radiation. New "bird-safe" instruments designed and made at SRP eliminated the problem. We learned which instruments required a low humidity environment. The need for additional offsite stations to monitor air and collect rainwater became apparent, and these were put in service in Allendale and Waynesboro, Georgia, soon after the preoperational survey ended. Similar stations were placed about 100 miles from SRP to assist in differentiating SRP releases from fallout.

Many other organizations contributed to the survey. The Corps of Engineers supplied soil samples from test borings, and the Bureau of Mines assisted in thorium analyses. The Philadelphia Academy of Natural Sciences and the Universities of Georgia and South Carolina collected and identified animals and plants. The Coast Guard helped to collect samples in the Savannah harbor. Instruments to measure environmental radiation were calibrated at the Medical College of Georgia. Local health officers assisted in collecting public water samples. The associations with the public health officers, universities, and the Philadelphia Academy of Natural Sciences that started during the preoperational survey were maintained and strengthened during the past 50 years.

Looking back, I'm surprised that none of the team members resigned during the survey. They slogged through swamps; side-stepped alligators; carried a snake-bite kit with an intimidating sharp razor blade; and suffered the hot, humid summers when working inside and outside. Those members with families tolerated inadequate housing. But the technical challenges of this first-of-a-kind activity and the thrill of being part of the atomic age, which was still new and exciting, evidently outweighed these conditions. They knew that the organized commotion of one of the largest construction projects ever undertaken in this nation would be an unforgettable experience. Another reason may have been that team members believed that in a small way, they were contributing to the security of the nation. These were times of considerable international tension. The U.S. was at war on the Korean peninsula, and the Soviets conducted their first nuclear weapons test in 1949 and in 1952 tested a thermonuclear device.

Why should this study made 50 years ago be considered significant today? My answer is simply this—the preoperational environmental survey inaugurated the long tradition of environmental stewardship that has served the Site and its neighbors so well. By focusing on environmental radioactivity, it helped to imbue in the institutional consciousness of the Site the importance of controlling releases to the environment. Several years after startup, management's philosophy regarding release of radioactivity was explained in the Congressional testimony of J.E. Cole, a director of Du Pont's Atomic Energy Division. In summarizing his statement he said, "It would seem tragic to discover in the year 2000 that improper confinement in prior years had made limited use of some of our water and land necessary, and this by an industry which was hailed with so much hope in 1960!"

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Biography

William Reinig served in the Navy during World War II. He received a Bachelor of mechanical engineering from the Polytechnic University. Before joining Du Pont in 1951, he was a health physicist at the Hanford Works and supervised the health physics group at Brookhaven National Laboratory's research reactor.

During Du Pont's operation of the Savannah River Site, he managed the Health Physics and Technical Departments. Under Westinghouse at SRS, he was Deputy General Manager of the Environment, Safety, Health, and Quality Assurance Division, and before his retirement in 1993, he was the principal scientist in that division.

He is past-president and a Fellow of the Health Physics Society. He was chairman of the American Board of Health Physics and a director of the American Academy of Health Physics. He is a consociate member of the National Council on Radiation Protection and vice-chair of Citizens for Nuclear Technology Awareness.

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High Sensitivity Measurements of Ultra-Low Amounts of Radioactivity in the Environment

Albert L. Boni

Abstract

Since the first water sample was taken from the Savannah River on July 26, 1951, continuing technological advances in high sensitivity measurement of ultra-low amounts of radioactivity are being made at the Savannah River Site (SRS). Notable achievements in this nuclear technology area have been recognized locally, nationally, and internationally. During the "Cold War" peak nuclear material production period, the capability achieved in high sensitivity radioactivity measurement technology demonstrated the resolve of the Savannah River Site to be good stewards of the environment. This is further demonstrated by the extremely low doses of radiation received by the surrounding population from Site operations, which are far below that from the natural environment.

Continued achievements in high sensitivity ultra-low radioactive measurement technology since the Cold War have created, in addition to site emergency response, new missions in nonproliferation international safeguards, national security against terrorism, nuclear smuggling, and state and local law enforcement.

It is these achievements at SRS in high sensitivity ultra-low measurement capabilities that make for a cleaner, safer, and more secure nation and world.

In the Beginning

The development of measuring ultra-low amounts of radioactivity in the environment began following the first water sample collected from the Savannah River in early 1951 by a core of Du Pont health physicists led by Bill Reinig. This was the first pre-operation environmental survey ever conducted prior to the construction and operation of a U.S. nuclear production or commercial facility (Bebbington 1990).

In conjunction with the Philadelphia Academy of Science and under the direction of Dr. Ruth Patrick of the Department of Limnology, river and the surrounding land environment monitoring stations were established out to 100 miles of the Savannah River Plant (SRP as it was then known) to routinely collect samples for radioactivity measurement. During the next one and one-half years (June 1, 1951 to January 1, 1953), about 6600 environmental samples were collected and analyzed.

By today's standards, only crude technologies existed in 1951 to measure radioactivity. These technologies included the analytical wet chemical separation of elements, light emitting phosphors (ZnS) for measuring alpha activity, and Geiger Mueller counters using a counting scalar to measure beta-gamma activity. By knowing the element separated, the radioactive isotope could be closely identified. Using a series of thin aluminum plates of known thickness (mass) placed between a separated element and the counter, an absorption curve could be produced to determine the energy of the activity and the radioactive isotope from its discrete beta energy emitted. This very tedious method required many technical analysts and rooms of counting equipment to determine the very low amounts of background radioactivity, which are naturally occurring in nearly all materials including the human body.

The Methods Chemistry and Radiation Physics Groups, composed of several chemistry and engineering scientists, were formed in the

Health Physics Department to improve upon these measurement methods to be more efficient, cost-effective, and become more sensitive to meet a rapidly growing concern of the not only local but the entire American public of the hazards of nuclear radiation. The arrival of atmospheric fallout from the first Soviet Union nuclear weapons tests and the increase in U.S. atmospheric testing demanded more advanced high sensitivity radioactivity measurement technologies. Such advances were necessary to maintain the health of our SRP and other Atomic Energy Commission (AEC) workers, U.S. citizens, and the world population and to monitor the nuclear threat posed by the former Soviet Union and other potential nuclear weapon proliferators.

Early Developments

Early high sensitivity measurements of ultra-low amounts of radioactivity used gamma spectrometry where gamma rays emitted by many radioactive isotopes are identified by the energy of the gamma ray or rays they emit. This is done by measuring the energy deposited in a scintillator such as sodium iodide (NaI) in crystal form and emitted as light energy converted to electrical pulses, which register the energy over a nominal scale of 10 to 3000 kilovolts. The spectrum made up of these peaks from a sample will identify all of the radioisotopes present that emit gamma radioactivity without having to perform wet chemistry quantitative elemental analysis. This method provided SRP environmental monitoring with a cost-effective and sensitive method to analyze thousands of samples relatively quickly.

By combining better methods to shield against background radiation from the natural environment, the first whole body counter was constructed as a room using 12-inch thick pre-World War II battleship armor plate. In 1959, a room was constructed as a box with a 5-ton steel door and lined with lead and copper plate to reduce low energy x-rays produced by cosmic interaction with the steel (Winn et al. 1986). The use of pre-World War II armor plate was to eliminate low background radioactivity due to fallout following the initial atmospheric testing of the atomic bomb. This state-of-the-art facility was used until the late eighties as the premier whole body ultra-low measurement technology for trace radioactivity that was naturally occurring or ingested by SRP employees.

This whole body measurement technology was also used in bioassay research studies to determine where radioactive isotopes would locate in the human body and how long they would remain to contribute to a person's lifetime dose. Such research contributed to the location and half-life of I-131 in the body, the half-life of Cs-137 in the body and its dependence on age (3-60 years of age) and gender (male or female), and the location and dose as determined using chest counting technology developments of plutonium inhaled and deposited in the lung.

With the development of liquid scintillation low energy beta measurement, tritium analysis for bioassay was simplified, from several days per sample to less than a day for 50-100 samples. SRP Health Protection scientists introduced the plastic bottle counting technology for bioassay tritium analysis, reducing the background and cost of quartz bottles while increasing sensitivity.

Field and laboratory concentration techniques were developed to achieve greater sensitivity and increase environmental monitoring sample throughput (see Figure 1). Large volumes of rain, river, and stream water were directly concentrated in the field and passed through specially designed ion exchange columns or liquid extractors, which increased the measurement of various radionuclides factors of 100-10,000 fold rather than using a 1 liter of water evaporation method. High volume air samplers were developed to concentrate particulate in air for volumes greater than 10 cubic meters per minute to track trace concentrations of atmospheric radionuclides. This technology was used in the late 1950s and 1960s to monitor atmospheric testing and its impact on the environ-

Figure 1. Early development of high sensitivity environmental collection and detection expertise

ment to empirically demonstrate the good stewardship of SRP in the protection of the environment and surrounding population. Following the atmospheric testing moratorium between the U.S. and Russia, this technology was used to detect (in the local CSRA area) atmospheric tests by the Chinese and the nuclear accident at Chernobyl. Ion exchange concentration of milk samples in the late 1950s and early 1960s was used to detect trace levels of I-131, Cs-137, and Sr-90 concentrations from fallout. The technique was so sensitive that the quality of milk could determine, by the naturally occurring K-40 content, who was diluting their milk with water. Another early technology was developed to increase detection sensitivity by improving quantity and geometry of the sample for direct counting by NaI gamma spectrometry, eliminating time consuming and expensive chemical separation techniques. In this technique, vegetation is compacted and dried up to 110°C to avoid loosing volatile iodine, which is measured.

Fallout was definitely a fortuitous benefactor to the development of the early sensitive measurement technologies for low-level amounts of radioactivity in the environment. Fallout provided the radionuclide tracers used in measurement research to achieve ultra low levels. Releases from SRP in the 1950s were extremely low and well below the AEC radioactivity release guidelines of the time.

Significant Achievements

Many significant achievements in the high sensitivity measurement of ultra-low amounts of radioactivity not only in the environment but in analytical measurement of radioactive and non-radioactive compounds, elements, and isotopes were developed at SRP. The further development of ultra-low background facilities to measure ultra-trace amounts of radioactivity resulted in an ultra-low level atmospheric gas measurement counting facility for tritium molecular forms released to the atmosphere (gas, oxide, organic) and various noble gases resulting from nuclear fission (weapons testing and commercial power reactors). The sensitivity levels now achieved are well below the 1 picocurie per cubic meter of air (less than 1 part in 1,000,000,000,000).

These achievements have also resulted in the construction of the only U.S. specifically designed underground ultra-low background underground radioactivity measurement facility (see Figure 2). The facility built in 1982 is a 9′ x 12′ x 7′ steel box constructed from 4-inch thick pre-World War II armorplate from the aircraft carrier Antiedim buried 50 feet underground surrounded by 4-6 feet of highly pure dense specular hematite ore. The facility contains no additional natural or man-made materials that could contribute radioactivity to the background. The facility is entered from a tunnel beginning 20′ underground from an above ground clean room facility where all of the information from the detectors within the underground shield is read out. This one-of-a-kind facility is used to support special site programs and work for other federal agencies. For example, NASA uses this facility to measure trace radionuclides in materials returned from space. The Department of Defense for Nuclear Safeguards also contracts to use this facility. The detection sensitivity of this facility is equivalent to less than one part in 1 trillion or the ability to find one penny in the U.S. national debt, about 5 trillion circa 2000.

The Tracking Atmospheric Radioactive Contaminants (TRAC) vehicle (see Figure 3) is another noteworthy achievement for measuring trace radioactive materials released to the atmosphere. The vehicle, built in 1982, is a single-body 22-ton vehicle with a chassis-mounted atmospheric sample collection and radiation real-time monitoring laboratory. Originally built for emergency response for unplanned SRS reactor releases of radioactivity, it now serves as a high sensitivity remote radiation measurements research and development laboratory and is available for emergency response to the Site and other federal agencies.

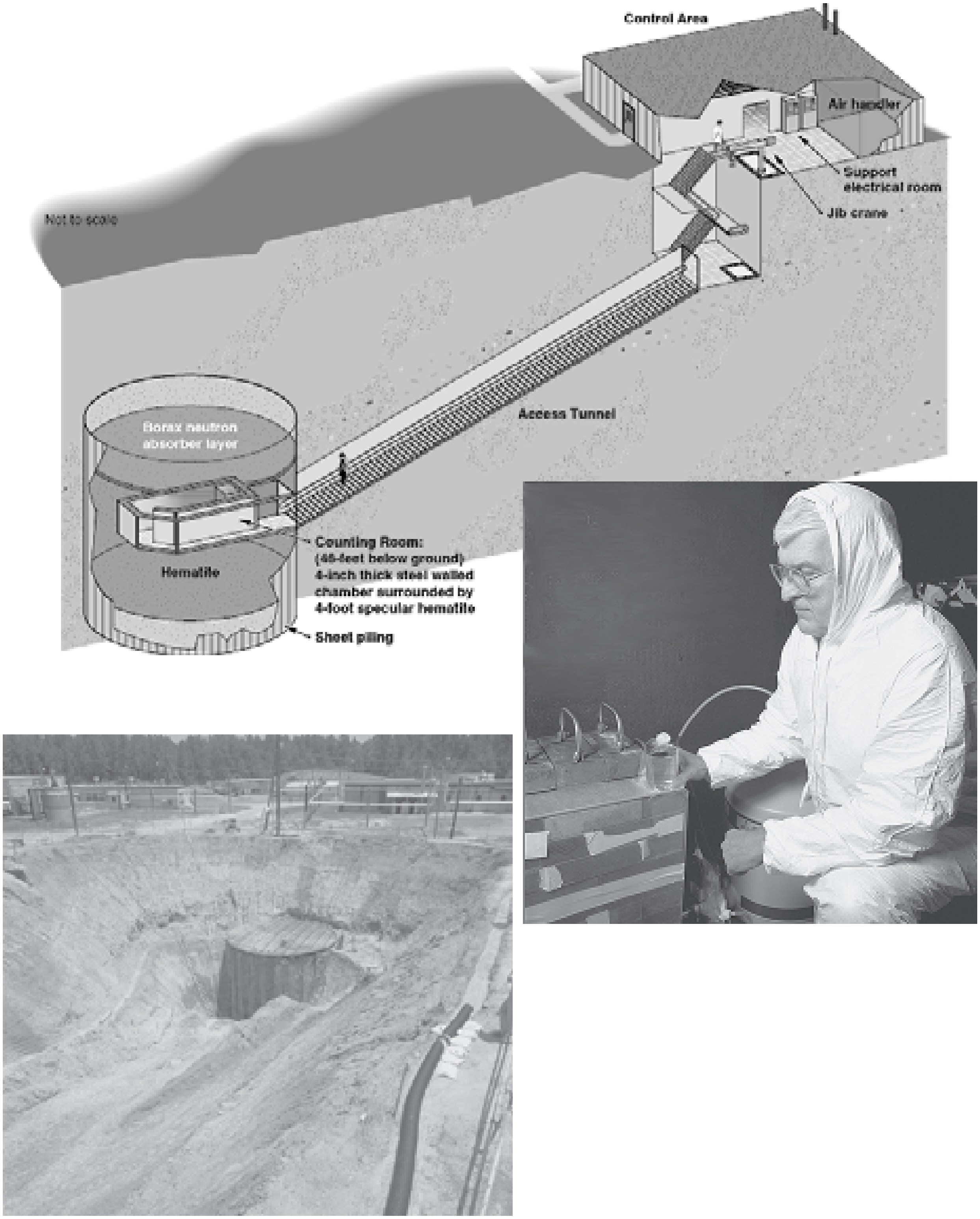


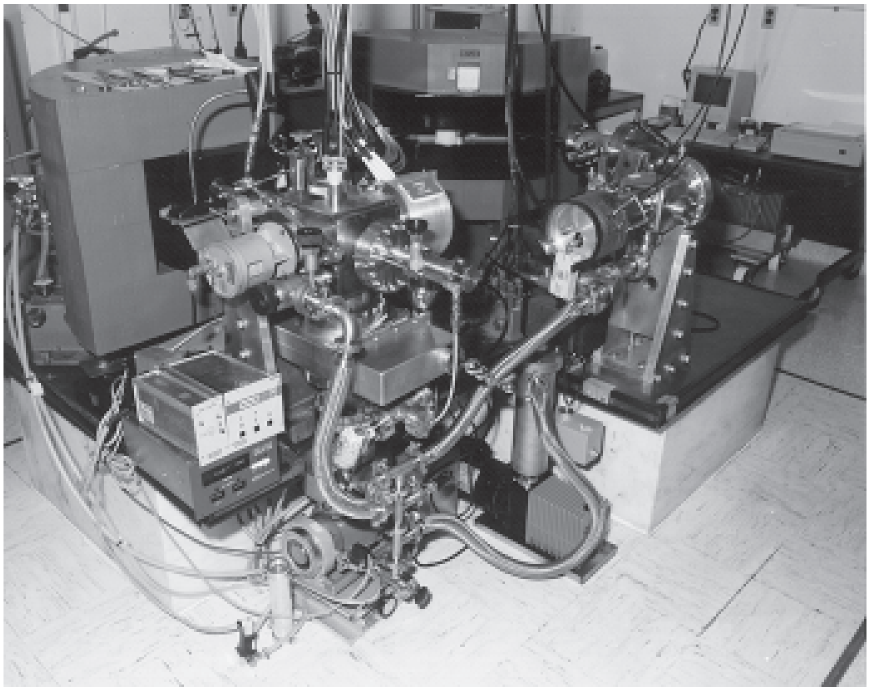
Figure 2. Highly sensitive counting and measurement—Underground Counting Facility

Figure 3. TRAC vehicle

The facility has a real-time gamma plume monitor for measuring a radioactive gaseous cloud, a high-volume 50-cubic-meter-per-minute atmospheric particulate filter system with real-time radioiodine and particulate radioactivity measurement detectors, and alpha surface barrier detectors to monitor actinides off-line. Detectors are also available to determine neutron activity levels at remote distances from a suspect source; when stationary, a portable high resolution Germanium detector for field sampling, and a low background liquid scintillation detector for off-line tritium oxide measurements in air or water. The vehicle is guided by geo-positioning satellite information system (GPS) and has local and distant radio communication. The vehicle can also be supplied with and periodically updated in real time with atmospheric plume transport maps through the SRTC Weather Center digital communication modem to aid in the location of radioactivity releases.

The measurement of ultra-low amounts of radioactivity by radiation measurement requires the emission of radiation by radioactive decay. Based upon their specific activity (curies per gram), many radioisotopes decay very slowly. Small amounts of these isotopes require weeks to measure by radiation measurement. SRS developed a highly sensitive mass spectrometric laboratory capable of measuring the actinides (uranium and plutonium) in a variety of environmental and bioassay samples that would require months by radiation measurement. In addition, this high sensitivity thermal ionization mass spectrometry technology measures the minor isotopes of uranium and plutonium that cannot be measured routinely by alpha spectrometry and identifies the source and age of the material, greatly assisting in contamination and unknown source investigations. The high sensitivity thermal ionization, time of flight secondary ion and x-ray fluorescence mass spectrometric laboratory (see Figure 4) has been greatly expanded to include developments in identifying the chemical signatures and morphology of individual micron size particles tracing them to their source and origin of formation. This forensic laboratory achievement has been recognized and routinely used for international nuclear safeguards, nuclear

Fourier Transform Mass Spectrometer (FTMS)



Thermal Ionization Mass Spectrometer (TIMS)

Figure 4. Forensic Science

smuggling, and law enforcement forensics investigations.

Numerous advances at SRS in the development of field and real-time radionuclide sampling, real-time radiation monitoring, and thermal remote sensing related to the measurement of small amounts of radioactivity in the environment have achieved national and international recognition (see Figure 5). Field sampling for atmospheric tritium chemical forms, streams and river water transport concentration of trace radionuclides, and the electrostatic high volume portable sampling of atmospheric radioactive aerosols developed at the SRS are widely used in international safeguards and national security programs.

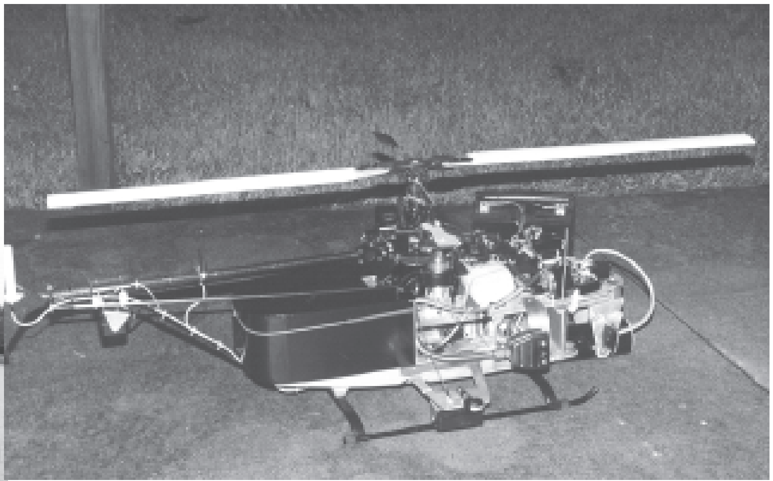
The high sensitivity measurement of small amounts of radioactivity in the environment is closely linked to the capability to locate in real-time sampling points and high probability of areas for collection and detection. This information is necessary to locate the source and measure the impact of the radioactive release on the environment and the surrounding population. SRS has developed one of the best meteorological and aqueous transport and dispersion real-time and forecasting centers in the nation (see Figure 6). Beginning in the 1950s, SRP used basic meteorological parameters, including wind direction, speed, barometric pressure, precipitation, temperature, and a standard relational graph to track unplanned releases. Today, an advanced three-dimensional model is generated from real-time meteorology data collected from 9 on-site towers and national and international meteorological data, updated every 3 hours. The SRS Weather Information and Display System (WIND) is recognized by the Department of Energy (DOE) and other federal agencies as a focal point for local and regional emergency response and in national and international nonproliferation safeguards and security circles. This achievement is responsible for the excellent emergency response capability and demonstrated good stewardship of the environment by SRS.

In addition to atmospheric transport, SRS has also achieved recognition for developing remote thermal sensing technology from U.S. multispectral satellites (see Figure 7). SRS is recognized by DOE as the ground truth (empirical ground measurement of temperature and meteorology for thermal sensing calibration) center for its newest multispectral thermal imaging (MTI) satellite to be launched in February 2000. SRS is recognized for its creative and innovative development of highly accurate measurement of the earth's surface temperatures. Such measurements are important to quickly determine with resolution on the order of square feet the source and magnitude of environmental pollution and natural disaster destruction over large areas of the earth's surface.

Several analytical high sensitivity measurement technology achievements include the use of C Reactor for real-time measurement of natural uranium and trace elements in environmental samples by real-time neutron activation coupled to prompt gamma, delayed neutron, and decay gamma emission. This was a nationally recognized analytical feat, which measured 100,000 samples over a 5-year period, for the National Uranium Resource Program to determine uranium deposits in the U.S. by taking environmental samples every 10 miles square. This amazing feat has never been duplicated. Because of expensive operating costs of the National Neutron Activation Facility in C Area, it was dismantled.

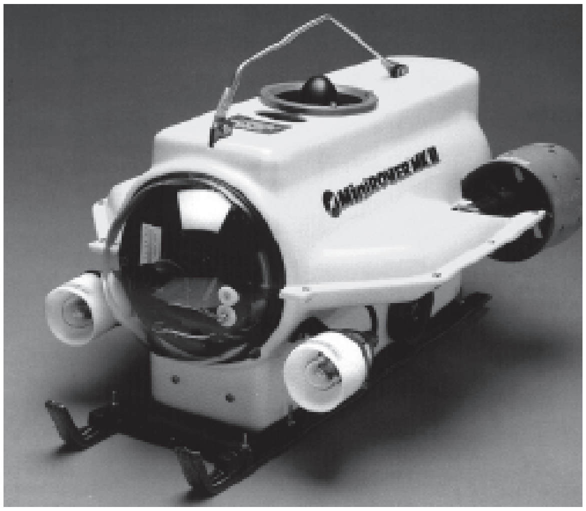
Another analytical high sensitivity measurement facility is the californium activation facility located in the Savannah River Technology Facility (SRTC). Although not the largest, it was one of the first such facilities and still remains in operation today. The facility houses a 1-Mg source of californium-252 in a large in-ground tank of light water and for shielding used an air pressure loading and removal "rabbit" (named for instant entry and exit of a small capsule upon initiation). The sample is housed in a small capsule called a rabbit, which after a selected time of irradiation by the

Borehole monitoring



Unmanned aerial vehicle

Underwater detector



Submersible

Figure 5. Unique field measurement systems

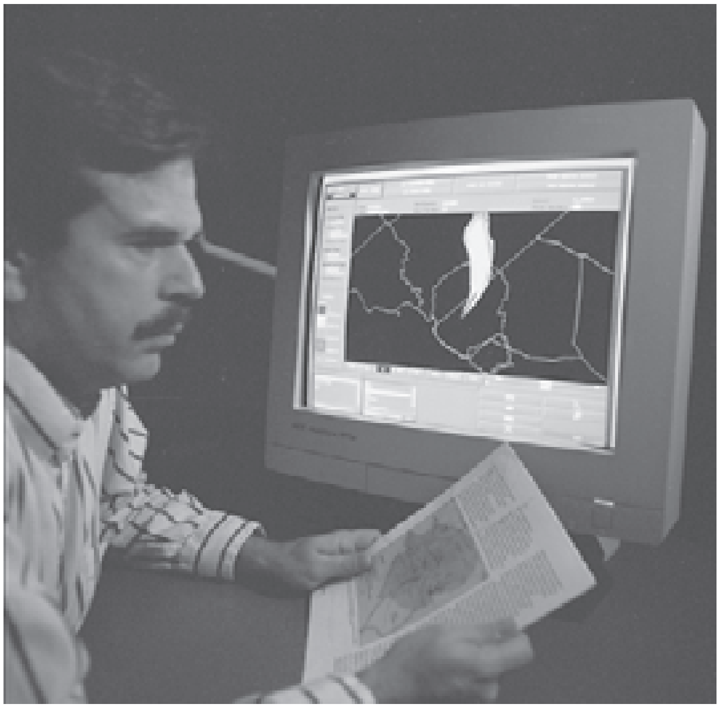


Figure 6. Meteorology

Figure 7. Remote sensing thermal imagery analysis

neutron source, is removed and counted immediately on a gamma spectrometric counting system. Californium-252 is a spontaneous fissioning isotope, providing an intense neutron source for activating stable isotopes and greatly increasing their analysis sensitivity through high sensitivity radiation measurement.

Missions in the 21st Century

Over the past 50 years since the first Savannah River water sample was collected, the innovative and creative development of high sensitivity measurement technology of small amounts of radioactivity has developed into a nationally and internationally recognized laboratory for ultra-trace radiation detection and measurement. Numerous health physicists, scientists, and engineers have contributed to this SRS technological achievement. Many are now retired, and a few remain; but, the creativity is carried on by new dedicated technologists. The technology of high sensitivity ultra-trace measurement of small amounts of radioactivity continues to grow and find new missions beyond the “Cold War” into new areas of nuclear nonproliferation, international safeguards, national security, and law enforcement.

International safeguards and nonproliferation efforts find SRS scientists at work in Iraq and other countries around the world conducting nuclear-related inspections (see Figure 8). These measurement technologies have also earned SRS/SRTC recognition as a nuclear forensic laboratory in support of the FBI in nuclear-related crimes such as international smuggling of nuclear materials and national nuclear terrorism. Although non-nuclear, the recognition of the Cold War high sensitivity technolo-

Figure 8. International nuclear safeguards: inspections and monitoring regimes to detect undeclared nuclear activities.

Figure 9. Regional law enforcement support (submersible vehicle)

gies has created a law enforcement mission to adapt these technologies to support South Carolina and Georgia state and local law enforcement agencies (see Figure 9).

Although the mission of high sensitivity measurement of ultra-low amounts of radioactivity in the environment may not be as large as nuclear materials production at SRS, it remains, even after 50 years and the end of the Cold War, important to the security of our site, nation, and the world. This technology continues to bring the most creative and innovative scientists and engineers to SRS, achieve recognition for SRS and SRTC, and continues to successfully provide the technology for a cleaner, safety, more secure nation and world.

Acknowledgments

It is impossible to acknowledge all the health physicists, scientists, and engineers who have contributed and those who are now contributing to this technological achievement. However, since the 1950s and the initial Methods Chemistry and Physics Methods Groups, there are early SRS health physics pioneers that deserve mentioning. They are C.M. Patterson, Bill Reinig, Ed Albenesius, Mac MacClaren, Paul K'burg, Jim Johnson, Al Boulogne, Henry Horton, Clarice Ashley, Eric Geiger, Harry Butler, Jack Hoy, Charlie Wright, Walt Marter, Ray Harvey, Gene Morris, Marshall Sanders, D.I. Ross, Ed Whitley, Sally Tucker, Bill Jacobsen, Bill Spell, Sally Leight, Worth Dalton, Dick Hawkins, Todd Crawford, Bob Taylor, Bill McMillan, Ed Miller, Ken MacMurdo, Larry Heinrich, and John Clark.

Present creative scientists include: Rob Addis, Al Garrett, Will Bowman, Burt Tiffany, Bill Winn, Justin Halverson, Bob Hochel, Jack Corey, Ray Sigg, Martti Kantelo, Ken Hofstetter, all our new young researchers, and all those technicians retired and present.

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Applied Environmental Technology Development at the Savannah River Site: A Retrospective on the Last Half of the 20th Century

Brian B. Looney

Abstract

Fifty years ago, the Savannah River Site (SRS) was built to produce nuclear materials. These operations impacted air, soil, groundwater, ecology, and the local environment. Throughout its history, SRS has addressed these contamination issues directly and has maintained a strong commitment to environmental stewardship. The Site boasts many environmental firsts. Notably, SRS was the first major DOE facility to perform a baseline ecological assessment. This pioneering effort, by Ruth Patrick and the Philadelphia Academy of Sciences, was performed during SRS planning and construction in the early 1950s. This unique early example sets the stage for subsequent efforts. Since that time, the scientists and engineers at SRS have proactively identified environmental problems as they occurred and have skillfully developed elegant and efficient solutions.

On a personal note, I am proud to represent the outstanding environmental scientists of the Savannah River Technology Center (SRTC, formerly the Savannah River Laboratory). Former employees such as Wendall Marine, James Fenimore, Henry Horton, Ed Albenesius, Bill Reinig and Todd Crawford, and current scientists such as Jack Corey, Al Boni, and Chas Murphy have served as role models and are my mentors. From these individuals, I learned that developing solutions to environmental problems requires honesty, simplicity, technical creativity, and hard work.

Introduction

The SRTC approach relies an interdisciplinary team of scientists—geologists, engineers, chemists, mathematicians, and others. The solutions developed by the team are based on focused environmental characterization followed by selecting and deploying cleanup technologies that are matched to the problem. Each technological advance is grounded in a clearly stated conceptual model and is developed and refined using the scientific method. Successful technologies always obey natural laws and often rely on natural processes or capabilities. These are the values that were instilled in me during my career in SRTC, and these are the values that I will try to communicate to you using a few examples below. Many of these technologies, consistent with the recent focus on partnerships with the community, have been transferred to the public for use in solving our nation's environmental challenges.

Anatomy of a Contaminated Site

Figure 1 depicts a conceptual diagram of a contaminated site that has impacted its surroundings—in this case, the underlying soil and groundwater. The three ovals—the source zone, the primary contaminant plume, and the dilute fringe—represent different portions of the impacted environment that each has a different character. The source zone contains significant contamination in concentrated and hazardous forms. The source zone can contain materials such as undissolved organic liquids (oils, fuels, or solvent), strong acids or bases, high levels of radiation, and/or toxic chemicals or elements. The second oval, the primary contaminant plume, is comprised of contaminated groundwater or vapor than carries pollutants at lower levels, but levels that still represent a potentially significant present or future hazard. The third

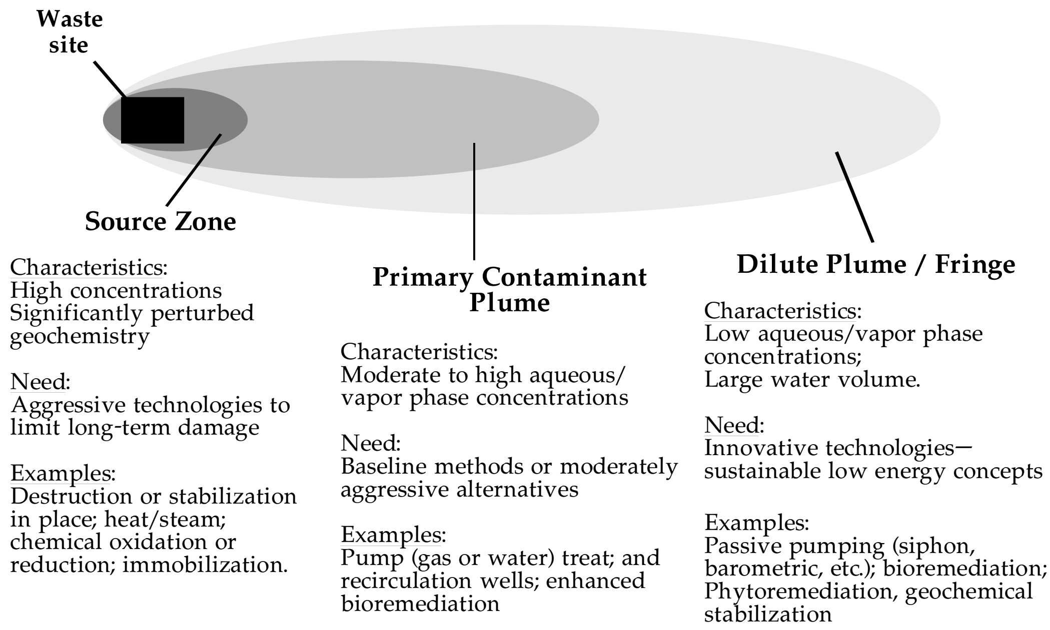


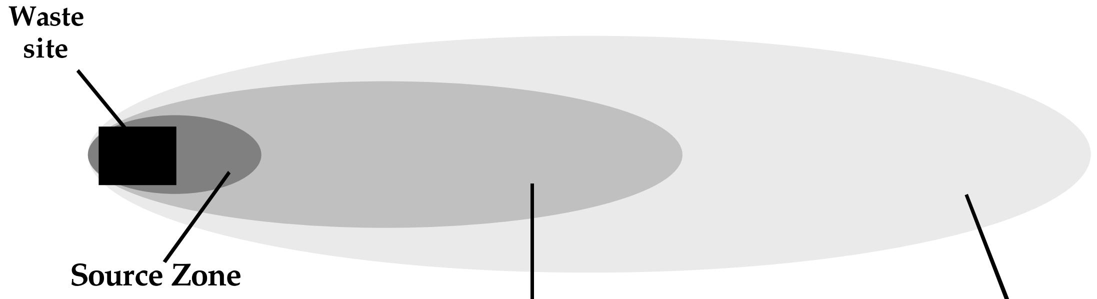
Figure 1. Anatomy of a contaminated site

oval, the dilute fringe, contains contamination at relatively low concentrations but in large volumes of water.

Efficient and effective environmental cleanup requires matching the character of the cleanup and stabilization methods to the character of the target zone of contamination. Thus, aggressive and relatively expensive methods are often appropriate for the source zone, classical pump-and-treat methods are often good for the primary contamination zone, and various methods based on natural processes are often best for the dilute fringe. Figure 1 identifies several example technologies that are appropriate for each of the ovals.

In Figure 2, I have extended this conceptual model by identifying the cost basis for the typical cleanup technologies. In the source zone, stabilization and removal methods are normally priced in terms of volume of soil or amount of contaminant in the treatment zone ($ per cubic yard, $ per pound, and the like). The reference source zone technologies require aggressive access and subsequent use of targeted energy or chemical reagents. It is clear that in the source zone it is important to characterize the site in such a way that the precise location of the source zone is delineated as carefully as possible. This approach will reduce costs by focusing energy or reagent to areas where they are needed. Equally important, however, is a desire to minimize of any undesired negative impacts (wasting energy, harming microbiological populations, etc.) associated with using aggressive remedies on regions without source level contamination. Similar to a doctor, environmental scientists should “first, do no harm.”

In the primary contaminant plume, treatment technologies are normally priced in terms of the amount of water (or vapor) treated ($ per gallon and the like). Thus, the goal of characterization is to define the flow directions and general plume structure to allow the most contaminant to be treated in the fewest “gallons”. Figure 3 illustrates an important final extension to our simplified conceptual model. This diagram of the primary contaminant plume at the SRS metals fuel and target fabrication facility (M Area) shows that contamination moves in response to many factors − contaminant release location and type, geology, sources and dis-



Source Zone

Costs:
$/lb contaminant or $/cu yd removal
Examples:
< $50-$100/cu yd or
< $100/lb for chlorinated solvents

Hot spot characterization
Reduces cleanup volume

Primary Contaminant Plume

Costs:
$/treatment volume (gallon/cu ft)
Example:
<$0.5-$10 / 1000 gallons

Zone of capture characterization needed, optimize extraction to reduce treatment volume

Dilute Plume/Fringe

Costs:
Operation and maintenance costs $/time

Mass transfer and flux characterization needed

Figure 2. Diagnosis and treatment of a contaminated site

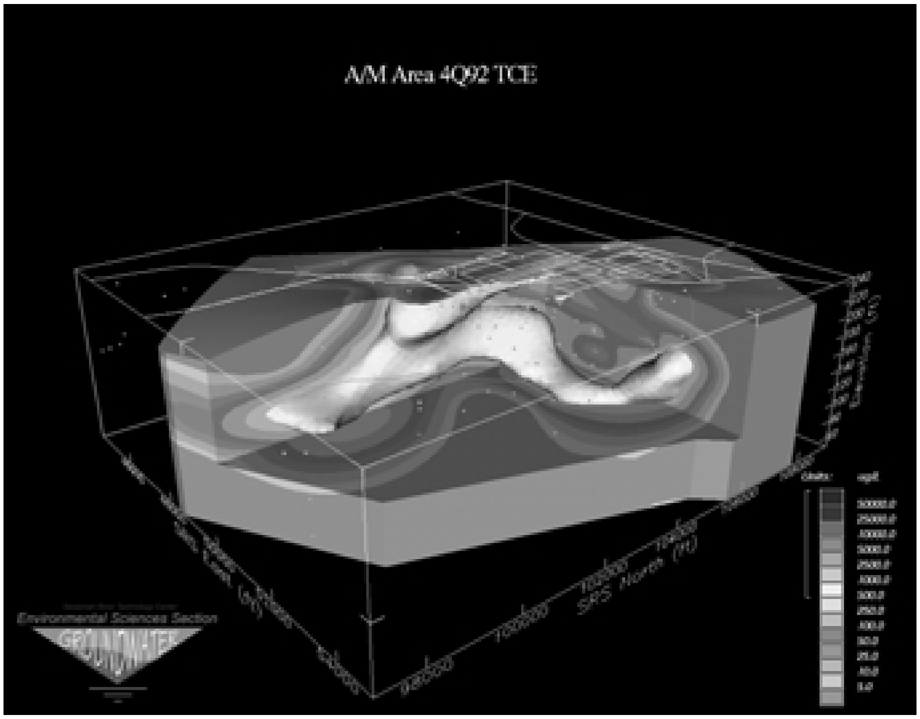


Figure 3. Cut-away diagram showing the 3-D structure of a real groundwater plume

charges of water, and many others. The resulting contaminated soil and groundwater zone occupies a complicated three-dimensional shape rather than the simple ovals that we began with. This complexity must be recognized when developing and implementing technologies for both characterization and clean up of the primary contaminant plume.

The dilute fringe contains low concentrations of contamination in large volumes of water. Thus, the best technologies for this zone are those that are priced in terms of time ($ per year and the like). To be successful, these technologies must rely on natural sustainable measurable processes. This class of technology has gained recent regulatory support under the terminology "monitored natural attenuation". For the dilute fringe, technology selection is biased toward understanding the contaminant destruction and stabilization capabilities of native species and natural populations. A second step is identifying engineering interventions, if needed, to maximize the performance and to assure that the attenuation process will operate for extended periods. A critical requirement for these technologies development of logical and cost-effective monitoring strategies.

The three zones depicted in Figure 1 are present at contaminated sites of all sizes. At a "mom-and-pop" gas station, the entire contaminated zone—all three ovals—might occupy a portion of a city block. At a large industrial facility like the M Area at SRS, the contaminated zone can extend over a few square miles. The size of a problem impacts how distinct the actions to address the different zones need to be. Time is also a factor. Concentrations change, as cleanup progresses, so that dilute fringe technologies become appropriate for polishing areas that were formerly at higher concentrations.

Above, I have outlined a conceptual description of a typical class of environmental problem. The description is simple and valuable. It provides clarity in defining what technologies are really needed. It helps us describe our clean-up plans to regulators and interested citizens. It encourages implementation of a suite of technologies—each targeting a problem that it is efficient and effective in addressing.

In the sections below, I highlight how this relatively simple conceptual model of the anatomy of a contaminated site can be the basis for improved environmental technologies. I have summarized examples of improved subsurface access methods, novel characterization techniques, and improved cleanup technologies for each of the ovals.

Source Zone Diagnosis and Treatment

As described above, it is critical to locate the concentrated and hazardous contaminants in the soil and shallow groundwater in the source zone. Data from most sites indicates that source zone contaminants accumulate in thin, highly concentrated layers—these layers can be only inches thick. Some contaminants concentrate near the point of release (many metals and radionuclides); others can move downward and concentrate at depth as they interact with hydrogeological features such as clay layers or the water table. The resulting challenge for characterization is to develop and use a strategy that defines these discrete intervals for a reasonable cost. Using traditional methods, namely drilling a few holes with limited numbers of expensive samples, has a high potential to miss the thin accumulation zones. While the samples may have a legal pedigree, such an approach does not efficiently support environmental decision making or engineering.

We have proposed a "toolbox" approach that uses technologies ranging from geophysics (looking at the reflection and transmission of energy through the soil) to traditional sampling. The heart of the toolbox for source zones, however, is a group of technologies (sensors and samplers) deployed by direct pushing, or insertion, into the ground (these methods have the generic name cone penetrometer [CPT], and trade names such as GeoProbe™ and SCAPs™).

These technologies directly address the problem of the geometry of the expected contaminant distribution. Using inexpensive sensors, CPT provides screening data throughout entire profile. The probability of identifying the thin accumulation zones is maximized.

Early CPT sensors were primarily used for describing geology and were developed for engineering and construction. These basic systems have been supplemented by an array of sensors that provide (as needed) electrical measurements, chemical measurements using spectroscopy or fluorescence (these use fiber optic lines to transfer light), direct viewing of the soil using cameras, and many different samplers to collect water, soil, or vapor. Several examples are described on Table 1. In each case, the technology is targeted at delineation of the high concentration source zone so that cleanup can be performed efficiently and safely.

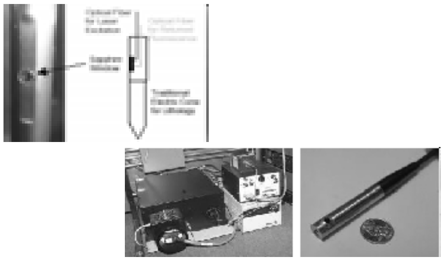
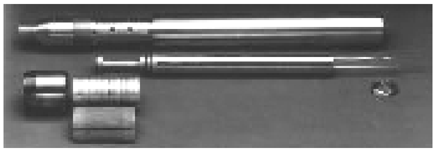
Characterization of a source zone is a necessary step toward the goal of removal and/or destruction. Appropriate classes of technologies to address source zone contamination include enhanced removal, *in situ* (or in place) destruction, stabilization, and barriers. These classes can be used alone or in combination. In collaboration with other DOE labs, federal agencies, universities, and industry, all of these source remediation technology classes have been tested and used (as appropriate) at SRS. With the exception of barriers, all of these technology classes require the "injection and mixing" of energy or treatment chemicals into the source zone. Energy-based technologies used at SRS include electrical resistance heating, radio frequency heating, and vitrification. A steam-based remediation, known as Dynamic Underground Stripping, is scheduled for full-scale use in 2000. Chemical-based systems range from shallow soil mixing units to reagent injection in wells. Figure 4 shows the operation of an example system in which Fenton's Reagent (hydrogen peroxide and reduced iron) was injected to destroy industrial solvents in a target zone about 150 feet deep.

Primary Contaminant Plume – Stepwise Improvement of the Baseline

This zone is characterized by the presence of contaminants at easily measured and potentially harmful. The contaminants in this zone tend to be somewhat mobile. As a result, baseline methods like "pump and treat" work reasonably well. Significant quantities of contamination can be removed (either as soil vapor or groundwater) and the contaminants treated at the surface using standard water treatment methods. Advancing the state of the art for this zone requires attention to large-scale plume geometry and incorporation of creative stepwise improvements in engineering. SRS has made several significant contributions that improve primary contaminant plume technolo-

Figure 4. Fenton's Reagent is added to a source zone to destroy NAPL in place. This project was a cooperation between SRTC and industry.

Table 1. Example CPT characterization technologies

	The cone penetrometer (CPT) and similar techniques such as the GeoProbe™ directly push sensors and samplers into the soil and shallow groundwater. This is a photograph of a CPT truck developed by DOD and DOE for testing new environmental characterization methods. The examples discussed below represents a collaboration among scientists from government agencies, industries, and universities.
	Fiber optic probes can be used with a CPT to measure chemicals and subsurface conditions. Spectroscopic measurements such as fluorescence (left picture) and raman (right pictures) can be related to chemical concentrations. Specialized sensors for a variety of uses have been developed, tested, and deployed.
	The Geo VIS Probe, a video system developed by DOD to be deployed with the CPT, is used to acquire magnified images of the soil and groundwater at the tip passes. The instrument consists of a CCD color camera, lens/focusing system, and an light-emitting-diode (LED) illumination system.
	A variety of samplers can be deployed using a CPT—samplers are available that collect liquids, vapors, or solids. Many systems allow samples to be collected without withdrawing the equipment from the hole. The cone sipper (left picture) is an SRTC-developed system used to collect vapor and liquid samples. In some cases, CPT can be used to install monitor in wells and other devices for long term use.
	Specialized measurements can also be made using CPT equipment. One example is the "permeability probe" developed by industry (Science and Engineering Associates).
	Creativity is a key to developing tools to find thin layers of contaminant accumulation. This fabric tube, modified by SRTC, can be installed in an open hole (installed by CPT or by drilling). The tube wicks oily contaminants—these release a dye and the stains on the retrieved fabric tube show the depth of contaminated layers.

gies. These contributions include improved depth discrete sampling devices (such as the Strata Sampler™ and the Cone Sipper™), improved data interpretation using 3-D imaging techniques, and successful deployment of innovative cleanup systems (recirculation wells and the like). I have summarized two notable contributions below—environmental horizontal wells and *in situ* bioremediation.

SRTC pioneered the use of horizontal wells for environmental cleanup. Environmental horizontal drilling has roots in oil and gas exploration and in shallow pipeline/utility installation. As depicted in Figure 3, the primary contaminant plume has a complicated 3-D geometry. The option of matching the geometry of a cleanup system to the geometry of the contaminant distribution using directional drilling, while simple in concept, represents a major advance. Horizontal and directionally drilled wells provide efficient access to contaminants, as well as a range of new and interesting engineering options (intercepting contaminants as they reach facility boundaries, cleanup underneath buildings, etc.). SRTC installed and tested two environmental horizontal wells in 1988—these wells represent the birth of the industry. Currently, SRS has nine horizontal environmental wells installed at several sites for a variety of uses. SRS research, combined with efforts of others, has resulted in growth of a mature and active horizontal environmental well industry and formation of a national technical and trade association.

SRTC innovative cleanup of the primary plume extends beyond optimizing geometry and improving access to the contamination. SRTC research has documented that natural microorganisms (bacteria, fungi, and the like) that are capable of destroying or stabilizing many pollutants are present in virtually all soil and groundwater systems. SRTC is recognized as a leading institution in developing and implementing methods to utilize this resource – putting these organisms to work for us. We "pay" them by adjusting the natural conditions and providing nutrients that are missing or limiting the rate of decontamination. In the case of gasoline and oil, the beneficial bacteria and other microorganisms consume the pollutants as a primary food source. To do this, they need oxygen, nitrogen, and phosphorus. Injecting air (oxygen and nitrogen) provides two of these nutrients. SRTC developed and patented a method of adding phosphorus to air so that all of the important nutrients could be added inexpensively. This technology, PHOSter®, has been widely licensed and is being used throughout the country to clean up sites ranging from "mom and pop" gas stations to large industrial sites.

SRTC has also pioneered technologies to clean up more challenging contaminants like industrial solvents. While these compounds are not directly used as food, we can add appropriate foods that encourage their destruction. Similar to PHOSter®, we developed a method based on adding air as the carrier. In this case, the air includes trace levels of natural gas and nutrients. The success of this technology has resulted in its licensing and use at a variety of sites across the country. The success of the bioremediation methods developed by SRTC is a testament to the scientific approach and conceptual model – that nature provides the basis for the best environmental solutions.

Dilute Fringe—Green Technologies

In the dilute fringe, even more than in any the other zones, the concept of putting nature to work for environmental cleanup central to success. Creative use of natural forces, natural laws, and site-specific conditions is the key to developing cost-effective solutions for low concentrations of contaminant in large volumes of water. Properly configured, tides, weather patterns, gravity, interfacial interactions, natural biological processes, and other basic forces, supply energy and mechanisms for contaminant destruction and stabilization. As discussed below, these processes can be inexpensive and effective. Importantly, the goal of dilute fringe technologies should be to reduce contaminant exposure (flux), to protect human and environmental health, and to monitor the performance of the protection in a direct and cost-effective manner. I have highlighted two SRTC-developed examples of the "green" technologies that are needed to address the challenging conditions in the dilute fringe—BaroBall™ and Geosiphon™.

The BaroBall™ is a remediation tool that uses variations in barometric pressure to extract contaminants from or inject fresh air into the soil. Without the device, wells screened above the water table inhale and exhale in response to the weather. Soil properties, depth, and other factors determine the amount of flow. The BaroBall™ is a simple check valve that uses a ping-pong ball to control flow. Consistent with the need for steady long-term cleanup of dilute fringe levels of contamination, the device provides a reliable performance with minimal use of using energy and minimal maintenance.

A similar creativity in using natural forces is embodied in the Geosiphon™. If left alone, contaminated groundwater moves steadily from its source to a discharge point near a river. Water is moving from higher head, or total pressure, to lower total pressure. The Geosiphon™ recognizes this reality and uses the simple concept of a siphon to exploit the pressure difference to our benefit. The system connects the contaminated groundwater to the river through a large pipe. Importantly, the system contains a treatment bed that purifies and detoxifies the water as it is being siphoned. To operate the system, the large pipe is primed and then the valves are open. Under the influence of gravity, the siphon extracts and treats the water without the need for a pump or pumping power (see Figure 5). The low concentrations in the dilute fringe result in a long life for the treatment bed and the overall system is conceptually appropriate for this zone.

As demonstrated by the use of ping-pong balls and siphons, it is clear that environmental technology solutions do not always need to be complicated. Particularly in the dilute fringe, simplicity and creativity are needed. SRTC is studying the potential role of plants and microorganisms near groundwater discharges to determine their potential for contributing to the solution. Dilute fringe technologies must be technically based and must be able to be monitored and documented.

Concluding Remarks

Similar to any large industrial facility, construction and operation of SRS resulted in many significant adverse environmental impacts. Nonetheless, it would have been easy to write this paper as a list of successes and statistics.

SRS has treated more than 3 billion gallons of groundwater and removed more than 800,000 pounds of contamination from soil and groundwater. SRS has completed or is actively cleaning up more than 300 of its 500 contaminated acres. SRS has been awarded 19 environmental technology patents and many national awards for its environmental accomplishments. SRS is committed to meeting its obligations under a wide array of environmental regulations—NEPA, RCRA, CERCLA, NPDES. The SRS Environmental Restoration Program has been the most active and successful program in the DOE complex in incorporating new technologies into its work to accelerate cleanup and reduce costs. SRS provides frequent and detailed public information on its environmental impacts. SRS was a charter federal facility designated as National Environmental Research Park. SRS is home to the preeminent ecological research center in the world—the University of Georgia Savannah River Ecology Laboratory and a major office of the U. S. Forest Service.

SRS applies creative, interdisciplinary approaches to developing inexpensive and robust technologies. The result has been a steady stream of effective products. SRTC pioneered the use of horizontal wells for cleanup, and has created and licensed important environmental samplers and sensors. Barometric pressure, solar energy, and microbiology all have been put to work. Going far beyond hatching new ideas, SRS is also recognized as a leader in developing and demonstrating new technologies. SRS has

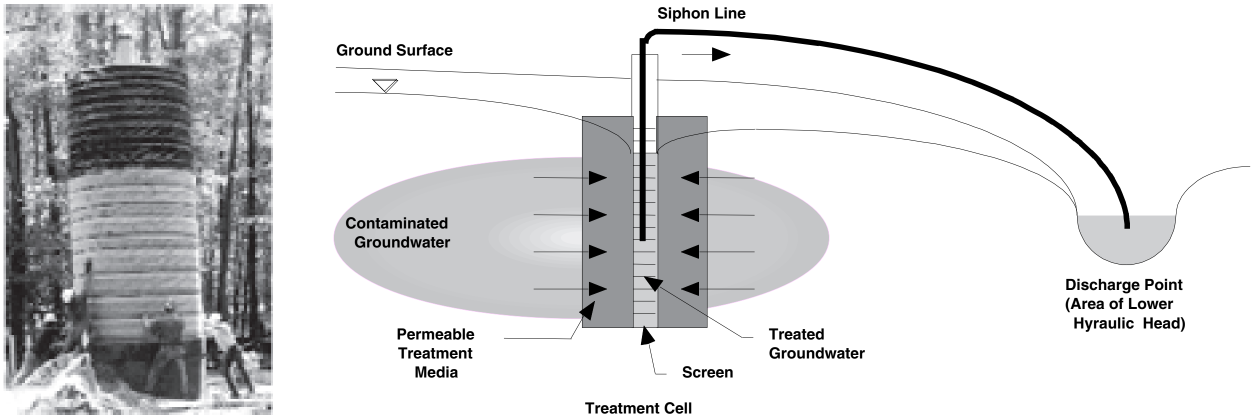


Figure 5. The SRTC GeoSiphon concept (right) and the installation of a GeoSiphon at SRS (left)

been instrumental in moving technologies, both those developed onsite and offsite, to field deployment and into widespread use in the private sector.

Lists of progress and accomplishments for each identified "waste site", as well as general chronologies of SRS accomplishments are widely available. I chose, instead, to provide my assessment of how SRS has achieved environmental progress—focusing especially on the technology contributions of the Site's scientists and engineers. We are committed to continue the fifty-year environmental technology legacy that has been entrusted to us.

Biography

Mr. Brian B. Looney is a senior fellow research engineer at the Department of Energy Savannah River Technology Center in Aiken, South Carolina. In this position for the past 16 years, he has coordinated development and deployment of environmental characterization and clean-up methods. He currently holds six U.S. patents and one foreign patent for environmental technologies. His interests range from basic science (geochemistry and the like) to engineering and implementation of cleanup methods. Mr. Looney's early research on environmental horizontal wells is a foundation for widespread use. He has been an integral part of SRTC teams developing innovative characterization methods, sensors, and samplers. He has supported recent programs targeting clean up of source zone contamination (using destruction and/or enhanced removal methods), as well as methods for dilute fringe contamination (barometric pumping and phytoremediation). He is active in national societies and participates in national scientific review and planning activities for the Department of Energy. He has many technical publications and is coeditor of a recent book on vadose zone science.

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Role of Microorganisms in the Operation of the Savannah River Site

Carl B. Fliermans

Abstract

Microorganisms are invisible to the naked eye, but their size belies their important role in nature and their role in the operation of the Savannah River Site (SRS). This contribution to the 50-Year Celebration of Excellence in Science and Engineering at the Savannah River Site details one of the microbial investigations that have provided greater insight into the versatility and applicability of our smallest allies to solve some of the greatest needs of humanity. The microbiological investigations at the Savannah River Site have opened new avenues for research into the interactions between the biosphere and the geosphere. The studies at SRS have shown the extension of the biosphere deep within the geosphere and that life may only cease to exist when temperature and pressure become inhospitable. These investigations have expanded our horizons about the habitats where microorganisms live and their ability to adapt and alter their selected niches. As we better understand the microbial niches around and under us, the sophisticated microbe continues to amaze its viewers, and in turn provides solutions to some of mankind's most pressing needs.

Proverbs 3:13 "Happy is the man that findeth
wisdom, and the man that
getteth understanding
14 For the merchandise of it is
better than the merchandise
of silver, and the gain thereof
than fine gold."- Solomon

Introduction

Microbiological investigations at the Savannah River Site have never taken on major mission status. It has been difficult for management to appreciate how invisible life plays a substantial role in the enormous science of big energy generation and nuclear weapon technology, but they have and they do. Microorganisms are important and affect all we do even in the nuclear industry. This is one of their stories as told by one of their friends. The narrative is of how subvisual life has influenced the science of subatomic particle physics. The reactors at SRS await their call. Environmental cleanup ensues, but the microbial world thrives as information flows from data defined by knowledge with a little wisdom of the microbial world around us.

In our family a clear message that has passed through the generations is that if one sees a turtle on a lamp post, one knows that the turtle did not get there by itself. It had lots of help. It is with this same sense of inadequacy and privilege that I have the honor of sharing one of the fascinating achievements at SRS in its 50-year history. Much of the data discussed in this contribution have been documented elsewhere (Fliermans and Balkwill 1989; Hazen et al. 1996; Massmann unpublished results).

When I arrived at SRP in 1974, Dr. Todd Crawford gave instructions to "set up a laboratory for microbial ecology". What a beautiful challenge. Little did he nor anyone else dream where that challenge would lead. Interests in the early days were of what microorganisms were able to do "to you" as opposed to the current emphasis at SRS as to what they can do "for you". Fresh from a post doc and studies on and in the extreme environments in Yellowstone National Park, I had eager excitement of how the microorganisms in natural thermal habitats compared to those in the man-made thermal habitats of the cooling water canals from the Site's nuclear reactors.

In the southeastern United States just a little thermal impact into already warm surface waters provides a perfect temperature habitat for those organisms that are capable of causing disease. With Ray Harvey and Lawrence Tilly as my mentors and J.J. Foreman as my right hand side kick, we began the adventure that continues. The journey serpentines through the intricacies of medical microbiology, consummating, in what has been described by the late Professor Rene Dubos, as a "very elegant marriage to microbial ecology" with the discover of the natural habitat of the bacterium that causes Legionnaires' Disease. That work has continued for the past 22 years and has been a vanguard in moving the understanding of microbial systems and interactions from data to information to knowledge with a little wisdom.

Microbiology of the Deep Subsurface

As thermal research became less of a DOE mission after the reactors were shut down, our interests began to shift towards understanding the ability and role of microbial systems in remediating contaminants left by the nuclear legacy in the groundwaters and terrestrial subsurface environments of the Site. There was but one problem. Extensive microbiological investigations had been confined to the upper few meters of the earth's crust. Until the beginning of DOE's Subsurface Microbiology Program in 1985 at the Savannah River Plant, scientists considered it unlikely that communities of microorganisms could inhabit the deeper sediments of the biosphere. In his seminal textbook Professor Martin Alexander of Cornell University, the foremost soil microbiologist of his day, stated, "THAT LIFE DOES NOT EXIST BELOW THE ROOT ZONE OF PLANTS" (Alexander 1977). Since the subsurface contaminants were relatively deep at SRS, and certainly below the root zone of plants where the conventional wisdom said "all" the microorganisms resided, such "dogma" had a dampening affect on any investigations that used microbial systems to clean up deeply affected aquifers and nonsaturated soils called the vadose zones.

If SRP were going to use microbial systems to affect the clean-up of hazardous and toxic wastes in the deep subsurface, then substantial microbial populations present in the subsurface would be required. Using state-of-the-art microbiological technologies, the Subsurface Program initiated at the Savannah River Plant focused on detecting microorganisms at great depths, establishing fundamental scientific information, including their ecology, and exploring their potential use in cleaning up contaminated deep terrestrial sediments and groundwater environments from energy and defense production activities (DOE 1988). Results from this program destroyed the traditional scientific concept of an abiological terrestrial deep subsurface. These investigations demonstrated that the terrestrial deep subsurface is a habitat of great biological diversity, and that activity does not decrease significantly with increasing habitat depth.

The enormous diversity of the microbiological communities in deep terrestrial sediments is most striking. Even at depths greater than 1000 feet, the number of microorganisms are greater than 10,000,000 bacteria per each gram of sediments. The organisms varied widely in their appearance and the way they transform or degrade a variety of organic and inorganic compounds. Regardless of the depth sampled, microorganisms were able to perform their traditional vital roles of recycling carbon, nitrogen, sulfur, manganese, iron, and phosphorus. Although the organisms were not of the same physiological types, each geological niche contained a basic cast of microbial players capable of these nutrient transformations. Such versatility was surprising, and contrary to conventional thinking about soil microbiology, because the deep subsurface was presumably a nutrient-limited environment where the driving force of life—photosynthesis and its products—are not abundant or absence.

The Microbiology of the Deep Subsurface Program has opened new avenues for research into the interactions between the biosphere and the geosphere. The biosphere now appears to extend a substantial distance into the geosphere and only ceases where temperature and pressure become incompatible with life. Recognition of the terrestrial subsurface as a microbiologically active environment applies to a variety of industrial and governmental concerns. It has influenced the planning for fossil fuel discovery, recovery, and storage; deep hazardous waste repositories; and groundwater remediation, storage, and retrieval. This recognition has provided new opportunities to produce and enhance biomedical and biological products and expanded thinking about extraterrestrial life. Knowledge about deep subsurface microbiology is likely to increase understanding of the transport and fate of groundwater contaminants, and it may offer new opportunities for *in situ* bioremediation strategies of deep groundwater and unsaturated vadose zone sediments. Microbial populations in these sediments are more active than had been expected from the scientific literature, and thus are likely to play a significant role in groundwater chemistry and geological processes. Additionally, these investigations have expanded horizons about the habitat of microorganisms and their ability to adapt to the parameters of the habitat and the ability of the microbial populations to alter their habitats. As we better understand the microbial niches around and under us, the sophisticated microorganisms we discover may help solve contamination problems, as well as provide useful products for humanity.

In Situ Bioremediation Demonstration of the Savannah River Integrated Demonstration Project

One of those applications of the Microbiology of the Deep Subsurface has been conceived and completed at SRS. The U.S. Department of Energy (DOE), Office of Technology Development, sponsored a full-scale environmental restoration technology demonstration at the Savannah River Site. The Integrated Demonstration Project, which began in 1989 in the M Area, enjoys national and international recognition for contributions to fundamental and applied research on innovative technologies for characterizing and cleaning soils and groundwater. The Integrated Demonstration has been described as the best bioremediation demonstration ever done. The primary emphasis of the subsurface remediation activities occurred in M Area at SRS, where the subsurface was contaminated by chlorinated solvents from the metal manufacturing facilities. This groundwater contamination resulted from surface spills and discharges from a variety of locations. The chlorinated solvents and degreasers are not unique to the Department of Energy facilities and are the most common soil and groundwater contaminants other than petroleum products. The two main solvents used in M Area were tetrachloroethylene (PCE) and trichloroethylene (TCE), man-made chemicals that do not occur naturally in the environment.

The Integrated Demonstration Program at SRS focuses on cleaning up soils and groundwater contaminated with volatile organic compounds (VOCs). To optimize resources, the project simultaneously evaluated and tested a large number of drilling, monitoring, characterization, and remediation technologies developed by SRS, other DOE sites, national laboratories, industries, and universities. During a single fiscal year (1992) over 44 different technologies were tested and evaluated. The principal remediation technology that this paper discusses is *in situ* bioremediation in conjunction with *in situ* air stripping. *In situ* air stripping was first demonstrated at Savannah River Site using parallel horizontal wells. These wells were placed in the subsurface, one below the water table and another above the water table. The initial *in situ* air stripping demonstration was successful in that it provided excellent characterization and monitoring data, which served as the background for the *in situ* biostimulation. Several collaborators had demonstrated in the laboratory the ability of a certain kind of bacteria to completely degrade or mineralize chlorinated solvents. These bacteria feed on methane and are naturally found in soils and aquifer material. The Integrated Demonstration Program injected methane mixed with air into the contaminated aquifer through horizontal wells and extracted from the vadose zone via parallel horizontal wells. This configuration has the advantage of simultaneously stimulating methanogenic activity in both the groundwater and vadose zone, and inhibiting spread of the organic plume.

Subsurface soils and groundwater adjacent to an abandoned process sewer line in M Area were found to contain elevated levels of the degreaser and cleaning fluid solvent, TCE (trichloroethylene). It has been estimated that roughly 3.5 million pounds of solvents were discharged to the subsurface in M Area. This area of subsurface and groundwater contamination was the focus of the Integrated Demonstration Program. TCE and PCE were first detected in the groundwater of M Area in 1981 where concentrations of PCE and TCE exceeded the drinking water level of 0.005 mg/L. This contaminant plume extended over an area greater than 1200 acres with a circumference of roughly 5 miles. The M-Area settling basin received solvents from about 1958 to 1979. Between 1958 and 1976, these solvents came from the 321-M facility. After 1976, solvents from Building 313-M were also discharged to the M-Area basin. Of the 2.1 million pounds discharged to the M-Area basin (1.9 million from Building 321-M and 220,000 from Building 313-M), it has been estimated that 84% was PCE, 15% was TCE, and 1% was TCA. Although the disposal of solvents to the M-Area basin was stopped in 1979, the basin continued to receive process effluents until 1985. The basin was certified as closed under RCRA in 1991.

PCE and TCE can be biodegraded or destroyed by naturally occurring microorganisms found in many soils and aquifer materials, including those at SRS (Fliermans et al. 1988). The primary biodegradation pathway for PCE and TCE involves bacteria that prefer to live in environments with low oxygen concentrations. These bacteria, called facultative anaerobic, prompt the sequential removal of chlorine atoms from the solvent molecules. PCE, with four chlorine atoms, is reduced to TCE and then to a compound with two chlorine atoms (DCE) and DCE is further reduced to vinyl chloride (VC), which has only one chlorine atom per molecule. Once the last chlorine atom is removed from the vinyl chloride, ethane is formed, which is a compound easily mineralized to carbon dioxide and water by a diverse group of bacteria.

These solvents were discharged into the vadose zone beneath M Area. The term "vadose zone" is used to describe the area between the ground surface and the water table. The water table identifies the area where the soil pores are completely filled with water. Above the water table (in the vadose zone) the pores contain both water and soil gases. The vadose zone is important to environmental restoration because essentially all groundwater contamination is derived from liquids initially introduced into the vadose zone.

Solvent contaminants occur in one of four different phases in the subsurface. A portion of the solvents will be attached to the soil particles; a portion will be dissolved in soil water; a portion will be in the vapor phase within the soil air; and a portion may be present as a separate liquid (non-aqueous) phase. Some of these phases can be removed more easily than others. While it is somewhat difficult to directly remove soil water or organic liquids from the vadose zone, it is relatively easy to remove soil vapor. In most situations, the only practical ways to remove these types of contaminants from most soils are the following:

- Physically remove the soils
- Induce chemical reactions that make the contaminants less toxic or less mobile
- Enhance biological reactions that degrade the contaminants or modify the contaminants to form a less toxic or less mobile phase
- Cause the contaminants to evaporate

The first approach, digging up the soils, is practical for relatively shallow contamination, although the excavated soils must generally be treated and replaced. Chemical modifications are difficult and are usually applicable only when soil concentrations are relatively high. The effectiveness of the biological reactions depends upon site-specific conditions but is often effective. The fourth approach, which is often termed soil vapor extraction (SVE), is usually effective for volatile organics compounds and, when combined with biological remediation, is one of the most common method for addressing subsurface contamination by volatile organic compounds such as TCE and PCE. Bioremediation can potentially enhance the performance of *in situ* air stripping as well as offer stand-alone remediation of contaminated sites. *In situ* air stripping is the mechanism where volatile organics are removed from the soil matrix by enhancing their transport from a liquid to a gaseous phase through increased air flow.

One of the ways a subsurface contaminant plume can be accessed is through drilling wells or boreholes. Contaminant plumes along a discharge pipeline, such as occurred in M Area, are generally elongated and elliptical in shape. Straight vertical wells that intercept the contaminant plume have a limited zone of influence, while horizontal wells, installed parallel to the plume, are much more capable of accessing the entire distribution of the contaminant plume. Dual horizontal wells parallel to each other were positioned in the subsurface of M Area in such a way as to be both above and below the contaminant plume thus manipulating the subsurface by enhancing the movement of gasses throughout the plume. The *In Situ* Bioremediation Demonstration was the first program to use horizontal wells for bioremediation. This technology was effectively demonstrated to recover groundwater contaminants for bioreactor conversions from deep or inaccessible areas (e.g., under buildings) and to enhance the distribution of nutrient or microbial additions in an *in situ* bioremediation. The horizontal wells are the base of the SRS Integrated Demonstration and are advantageous over conventional vertical wells for bioremediation nutrient delivery techniques. The increased surface area of the horizontal wells delivers more nutrients, recovers gas and water easier, and minimizes clogging in geological formation being remediated. The principal nutrient supplied via the horizontal wells in this test was methane in air, at a low concentration of less than 4%. The reason methane was added was because the microorganisms that degrade the chlorinated hydrocarbons are methanotrophic bacteria. That means that as part of their diet they get energy from eating methane as well as the carbon in carbon dioxide and methane. This is done through a group of enzymes called methane monooxygenases. These enzymes are somewhat sloppy in that they have a difficult time telling the difference between methane and TCE or PCE. Thus the strategy was to stimulate these particular bacteria *in situ* by giving them methane, but then forcing them to change their diet to effectively eat the PCE/TCE, and thus strip off the chlorines.

The lower horizontal well was an efficient delivery system for gases throughout the contaminated region. A vacuum was applied to the upper well in the vadose zone to encourage air/methane to move through the upper saturated zone and lower vadose zone, inhibiting spread of the contaminant plume. Air/methane mixtures have been demonstrated to stimulate selected members of the indigenous microbial communities capable of degrading TCE. Extensive characterization of monitoring wells and periodic sediment borings were used to measure the response of the soil and water following injection of air/methane. In addition, offgas from the upper horizontal well was assayed for methane, total volatile organic carbon, TCE, PCE, and potential breakdown of TCE/PCE (e.g., DCE, VC, and carbon dioxide).

Initially, 1% methane with air was injected continuously into the lower well. However to ensure process optimization (i.e., to further stimulate the indigenous microorganisms to enhance their biodegradation rates and efficiencies), the injection protocol was altered for subsequent campaigns. At 3-month intervals during the 14-month demonstration, the data were examined by an expert panel, and the final test campaign was developed for use:

- Air injection alone for the upper well at 249 scfm
- Air alone injection was added at 200 scfm in the lower well
- Injection with 1% methane/air in the lower well
- Injection with 4% methane/air in the lower well
- Pulsing 4% methane/air in lower well
- Pulsing 4% methane and continuous injection of nitrous oxide at 0.07% in air and triethyl phosphate at 0.007% in air into the lower well

During the test period, the flow and vacuum conditions of the extraction system have remained constant with a flow of 240 scfm and 7.6 inches of mercury, respectively. VOCs in the offgas were composed entirely of TCE and PCE, while the overall VOC concentrations started 10 times higher and declined rapidly over the next 5 days, and stabilized during the 14 months of the demonstration. Comparison of VOCs in pre-and post-test borings support this observation since sediment concentrations decreased by more than 30%. Interim borings at four holes done at the end of the 1% methane injection also reveal a further 50% decline of VOCs in the sediment. Indeed, few of these samples had detectable levels remaining.

Air injection (200 scfm) seemed to have little effect on the extraction efficiency. Methane injection at 1% and 4% had little effect on extraction efficiency of offgas quality, though overall there was a small decline in VOC concentration over time for both operating campaigns. In addition, the ratio of TCE/PCE significantly and consistently declined over time. This observation is consistent with knowledge that methanotrophs will degrade TCE, and to a lesser extent PCE, and that PCE is degraded at a slower rate by syntrophic anaerobes.

To attempt to optimize the methane-eating bacteria to favor TCE/PCE use, methane was pulsed-injected into the system. Pulsing methane significantly decreased VOC concentration in the extraction well. When the methane was injected again for 5 days after air-alone injection, the VOC concentration increased, but declined again as soon as this pulse was stopped. These observations coincide with the understanding of competitive inhibition (i.e., when high biomass was achieved then the methane is withdrawn, and more contaminants were degraded, since there were more enzyme-active sites available). In addition, it appeared that the long interval pulsing decreased methanotroph density during the first 6 weeks of the pulsing campaign. During the subsequent 6 weeks, the short-interval pulsing increased methanotroph densities. Carbon dioxide concentrations from the extraction well suggest an upward trend beginning 2 to 3 weeks post-air injection start-up. This may indicate increased microbial respiration in the subsurface caused by the air injection.

There was also a striking positive correlation between VOC concentration in vadose zone soil gas and CO_2 concentrations. After VOCs disappeared, the CO_2 concentration subsequently declined. When new VOCs moved into the area, the CO_2 concentrations subsequently increased until after the VOCs have declined again. Since pulsing began, vadose zone concentrations declined significantly and then increased in some wells. Since nitrogen and phosphorus (N&P) injection began, the concentration of VOC in all vadose zone wells declined dramatically, more than 90%. This again supports the theory of competitive inhibition and nutrient limitations discussed above. More than 108,206,345 scf of air were injected during this test. As expected, even though more than 1,392,774 scf of methane were injected into the subsurface during 53 weeks, only trace quantities of methane were detected in the extraction wells or any of the vadose zone piezometers during the 1% methane injection campaign (i.e., most, if not all, the methane injected was consumed by the TCE-degrading microflora). Simultaneous injection of helium as a conservative tracer has shown that more than 50% of the injected methane is being consumed.

Groundwater monitoring has shown that methanotrophic bacteria increased at the rate of one order of magnitude every 2 weeks since 1% methane injection began. However, increases substantially slowed and began declining slightly. This change coincides with reduction in nitrates in the water off these wells. Several other measures of microbial activity and abundance have shown a similar response to nitrates. After the 4% methane injection started, (8/5/92) methanotroph densities continued to increase. The wells showing the greatest decrease in TCE/PCE concentrations have experienced as much as a five order-of-magnitude increase in methanotrophs. These same wells have also shown increased concentrations of chloride in the water, an aerobic biodegradation end product for TCE. Stimulation of biodegradation activity by the indigenous microflora appears to have been great during the initial phase of the 1% methane injection. After 2 months of the 4% methane/air campaign, it appeared that the methanotrophic population was further stimulated, but the nitrogen-fixing bacteria were inhibited, causing severe nitrogen limitations. However, wells farther away from the injection point showed significant densities of methanotrophs and for the first time the concentrations of TCE/PCE either remained the same or declined slightly. The 4% methane injection appears to have inhibited nitrogen-transforming bacteria; therefore, we began the pulsing campaign, which initially consisted of air injection alone for 5 to 14 days, followed by injecting 1% methane for 4 to 5 days. It was believed this would reduce competitive inhibition of the methane and TCE for the same enzyme and reduce the inhibition of nitrogen fixers shown to be stimulated by air injection alone. Pulsing caused significant increase in nitrogen-transforming bacteria, a decrease in TCE in well water as well as the vadose zone, and a decrease in methanotroph densities. On December 11, 1993, the short-pulse interval campaign of 8 hr of 4% methane every other day was begun. The final campaign (January 25, 1993) included pulsed injection of methane and continuous injection of nitrous oxide at 0.07% in air and triethyl phosphate at 0.007% in air. The decision was based on enrichment and mineralization studies. It was felt that this last injection would overcome both N&P limitations and allow higher biomass and higher degradation rates of TCE achieved by the methane-stimulated subsurface bacteria. Since the N&P injections, the densities of the methanotrohic bacteria in the water have increased while the TCE concentrations in the vadose zone and water has declined.

The vapor extraction systems at M Area have been effective in terms of the amount of contaminant that has been recovered from the subsurface and the new methodology to allow such transformations. Operating costs indicate that bioremediation used in the *In Situ* Bioremediation Demonstration is far more cost effective (less than 50%) of the cost to remove a pound of contaminants via the groundwater extraction system. However, the total amount of contaminant that has been removed by the vapor extraction system is still a small fraction of the amount that was likely discharged to the subsurface. While the total amount removed via vapor extraction system (roughly 250,000 pounds) is less than 10% of the estimated amount that was discharged to the subsurface, the *In Situ* Bioremediation technology significantly enhanced the tools available to remediate deep subsurface sediments.

These investigations have demonstrated that the gaseous nutrient injections stimulated indigenous soil bacteria to degrade TCE and PCE without risk of forming potentially harmful daughter products or biofouling the remediation wells. One begins these investigations with a hypothesis that requires the collection of data and advances the collection of data about what microorganisms are present in a particular habitat (i.e., methanotrophs). The use of such data allows one to obtain meaningful information as to what these organism can do under real world conditions to address a particular problem. The next step in moving from data to the acquisition of wisdom is how to use the information to obtain knowledge. Once the information on the effectiveness of methanotrophs had been established, this information was linked with data and information from other disciplines (geology and engineering) to advance information into the application of that information towards knowledge. The application of that knowledge needs to be done with insight that provides a little wisdom in solving the problem of subsurface bioremediation. This project demonstrated not only the feasibility but also the effectiveness of *in situ* bioremediation of groundwater and sediments contaminated with chlorinated solvents. Indigenous microorganisms were stimulated to degrade TCE, PCE and their daughter products *in situ* by adding selective nutrients to the contaminated zone. *In situ* biodegradation is a highly attractive technology for remediation because contaminants are destroyed, not simply moved to another location or immobilized. Such technology decreases costs, risks, and time while increasing efficiency and public and regulatory acceptability. Bioremediation has been found to be among the least costly technologies in applicable situations.

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Biography

Carl B. Fliermans is a microbial ecologist with a doctorate in limnology and microbial ecology from Indiana University. Following postdoctoral work as an National Institutes of Health Fellow at University of Minnesota, Mr. Fliermans joined Savannah River Site in 1974 and established a microbial laboratory designed to address and solve environmental problems from both a basic research and applied perspectives.

Techniques developed during Mr. Fliermans' tenure at SRS has allowed his laboratory to discover the ecological niche of Legionnaires' Disease Bacterium and provide ecological research instrumental in the identification and control of the bacterium.

Lost Lake Found—Restoration of a Carolina Bay Wetland

Lynn D. Wike, F. Douglas Martin, and John B. Gladden

Abstract

Lost Lake is a Carolina bay wetland located on Department of Energy's Savannah River Site (SRS) near Aiken, South Carolina. Before closing access to SRS to the public, Lost Lake had been drained and planted as part of an agricultural field. In the 1950s, Lost Lake was allowed to refill and to return to its function as a wetland, but it was severely impacted by SRS operations. In 1991, Lost Lake was drained again and restored by removing and replacing contaminated sediments and through soil treatments. Studies of the amphibian populations before and after the restoration effort indicate that recovery is extensive. In addition to serving as an experiment in restoration techniques, this wetland has served as a teaching laboratory for graduate and undergraduate students in local colleges and universities.

A shimmer of light reflecting among stands of pines, dog fennel, and blackberry canes is the first glimpse one gets of Lost Lake. That glimpse comes unexpectedly considering the adjacent and surrounding area. Travelling past the closed M-Area seepage basin, numerous monitoring well heads, air strippers, injection wells, and other industrial paraphernalia and clutter associated with cleaning up a RCRA site, the last thing one would expect rounding a bend in the gravel service road would be to come upon a Carolina bay. Even more interesting is that this wetland is perched in the dry sandhill area with no obvious link to any water supply. Of course if you were an ecologist living and working in the southeastern United States, it would be exactly what you would expect and in fact, would be seeking. Actually, Lost Lake came by its name through just those circumstances. In the early 1970s, Drs. Whit Gibbons and Rebecca Sharitz of the Savannah River Ecology Laboratory (SREL) were looking for an unnamed Carolina bay near where new SREL facilities were to be built, but having forgotten the map, had no success (Gibbons 1990). Upon returning from the field, they named the elusive body of water Lost Lake. There is an irony in how that name was so historically correct and at the same time prophetic of the future of Lost Lake. Historically correct, because in 1950 when the Savannah River Site was closed to the public, Lost Lake had indeed been lost for quite some time. It did not exist except in the profiles of its rich wetland soils visible only from the air. The area where Lost Lake had previously existed, and exists today, was an agricultural field in 1950. Aerial photographs from the late 1930s and early 1940s distinctly reveal the outline of Lost Lake within the local fields and show the ditch used to drain the water from the wetland, allowing its use for agriculture. No one knows how long Lost Lake had been gone, but with closing the site and stopping maintenance on the drainage ditch, Lost Lake slowly returned to its former function. The SREL scientists' name for the bay was also prescient in that it inadvertently, but accurately, predicted the future disappearance of this Carolina bay in both the figurative and literal sense. Before we examine the known history of Lost Lake, let us examine exactly what makes a Carolina bay what it is.

Carolina bays are a unique form of wetland found on the southeastern U.S. coastal plain from Virginia to Florida. All Carolina bays are naturally occurring shallow depressions of interstream areas that share at least some of many characteristics. These characteristics include an age of at least 30,000 years, elliptical or ovoid shape with NW to SE orientation of the long axis, low sandy marginal rims with greatest development on the SE margin, substrate of either clays and silts or organic peat, and hydrology varying from seasonal to continual inundation (Schalles et al. 1989). Sizes vary greatly from one or only a few tenths of an acre to the size of Lake Waccamaw in North Carolina. The surrounding watershed determines hydrology of Carolina bays because they have no natural inflowing or outflowing streams. Anthropogenic influences, either directly by ditching, or indirectly by altering the surrounding watershed, have pronounced effects on the hydrology of Carolina bays. The fate of Lost Lake, that of being ditched and drained for agricultural use, has been the same for thousands of bays throughout their range, including many of the hundreds of bays found on SRS. Carolina bays exist as islands of water in the often xeric upland interstream areas. As such they provide important diversity of habitat and available water, forage, and breeding areas for a wide variety of organisms. Nearly all of the bays on SRS are mineral substrate, but they support a wide variety of vegetation types, depending upon their size and hydrology. Vegetation can vary from open herbaceous bays that remain wet all year to closed canopy forested bays that are wet for only a portion of the year. Some bays can remain dry for several years depending on climatic conditions. This varying hydrology has a large role in determining what type of plant community is found in any specific bay. The gradient from wet to dry from the center to the rim of bays tends to zone the vegetation communities. These zones vary, and 17 different herbaceous zones alone have been described, as many as 6 in a single bay (Schalles et al. 1989). Invertebrates, although not widely studied, can be abundant and diverse, depending upon the specific bay and its hydrology. Vertebrates have been studied extensively at several bays with amphibians being the dominant taxa. Over 30 species of amphibians and reptiles have been noted in a single bay (Gibbons 1970) and amphibian productivity can be very high because these isolated wetlands are often the only landscape feature available for amphibian reproduction in a relatively large area . Fish do not generally play a large part in the vertebrate fauna simply because the are not present in most bays. Bays that receive flooding from other sources containing fish may develop populations, but the varying hydrology and periodic drying will often eliminate any fish community. Carolina bays also serve as water sources for a large variety of terrestrial organisms and wildlife.

In the early 1950s, after SRS had been closed to the public and Lost Lake began to function as a wetland again, two important things happened around the bay. First the entire area, except for the existing 8-acre hardwood stand south of the bay, was planted in loblolly (*Pinus taeda*) and slash (*P. elliotti*) pine. The second and more serious event was the construction of the M-Area fabrication facility and its related support facilities. One of these supporting facilities was a settling basin that received effluents containing solvents and various salts of heavy metals. Lost Lake is downslope from this settling basin, and, on those occasions where the basin overflowed, these same toxicants ended up in Lost Lake. In the 1970s, Lost Lake was so heavily impacted by these substances that emergent vegetation, such as cattails or water lilies, and submerged vegetation, such as water celery, bladderworts or coontail, were completely absent. Despite this level of contamination, amphibians continued to breed in Lost Lake, perhaps because there was no place else to go. In 1977-1978, Steven Bennett and other Savannah River Ecology Laboratory investigators conducted one of the earliest examinations of the relationship of forestry practice and amphibian community structure at this site. Their study was designed to determine terrestrial activity, relative abundance, and diversity of amphibians in the three forest types surrounding Lost Lake (Bennett et al. 1980). This study was one of the first in North America to examine the roles of forest management practices in reptile and amphibian community structure. Lost Lake, at the time of Bennett's study, was so polluted that it did not support either emergent or submerged aquatic plants but amphibians were still breeding there. Remarkably, there were 5 species of salamanders and 11 species of frogs and toads captured during that study. Bennett reported that, while the two types of pine and the hardwood forest had the same species of terrestrial amphibians, the hardwood forest yielded approximately 50% more individual amphibians than either pine forest during both study years.

The dying Lost Lake was destined to be lost again and to be reborn. In 1990, a closure plan for the M-Area settling basin near Lost Lake was developed which included, in addition to closing the basin, restoring Lost Lake to a "natural wetland system" (Gladden et al. 1992). The Department of Energy (DOE) established a task team to develop a strategy and approach for the restoration activities at Lost Lake. The team included members from DOE, Savannah River Forest Station (SRFS), Soil Conservation Service (SCS), Savannah River Ecology Laboratory (SREL), and several organizations within Westinghouse Savannah River Company (WSRC). The committee was chaired by the Savannah River Technology Center (SRTC). Lost Lake was drained, and the surrounding vegetation within a minimum radius of 50 meters was removed (see Figure 1). All vegetation from the removal action was burned, and the residual ash and contaminated sediments were dug up and moved to the settling basin and compacted. Removed sediments were replaced with "clean" soil, and the bay was divided into eight segments, each of which received one of four different soil treatments. The bay was allowed to refill and aquatic vegetation was planted. Over 150 individuals of 10 different species of woody vegetation were also planted in the cleared area around Lost Lake. Before restoration, Lost Lake had a surface area of approximately 5 acres. Based on a 1996 aerial photograph, the current surface area is approximately 16 acres. This seems like a huge change in surface area. However, in Carolina bays, fluctuation in water surface area is normal; and the range of fluctuation for any given bay may be unknown, though the sandy marginal rim probably outlines the greatest surface extent possible.

From 1993 through 1996, we studied the amphibians and their recolonization of Lost Lake in an effort to assess the success of restoration. Because the amphibian populations colonizing the wetland inhabit or migrate through the three adjacent forest types, we were also able to reevaluate the relative abundance, diversity, and fluctuations of the populations in each of the three forests and to compare our results with Bennett's studies from before the restoration. Like Bennett, we found that amphibians were more abundant in the hardwood stand than in either managed pine forest. Also, of the 16 species Bennett captured, we caught 14. The gray treefrog, which Bennett captured, we did not capture, but one male was heard calling from near the shoreline during our study. The other species Bennett caught that we did not, the dwarf salamander, may not be able to recolonize Lost Lake for years, as its preferred habitat is wet leaf litter near the shoreline. Until the trees and shrubs grow back along the shoreline, there may not be suitable habitat for this species, which is otherwise common on SRS. Because Bennett only sampled in July and August, he did not capture several species that we caught in our winter and spring sampling. (For a list of all species captured, see Table 1.) Despite the good news that almost all prerestoration species are present once again, one point that may be of concern is that densities of amphibians in this area are now less than one fifth of what they were during Bennett's study. Whether this decline is the result of removing shrubs and trees near Lost Lake during restoration activities, the generalized global decline in amphibia, or due to unrelated events is unknown at this time. Although future studies may contribute to the solution of this puzzle, no research is currently being conducted, and none is budgeted for the near future.

The recovery of Lost Lake has been fertile ground for both ecological research and educa-

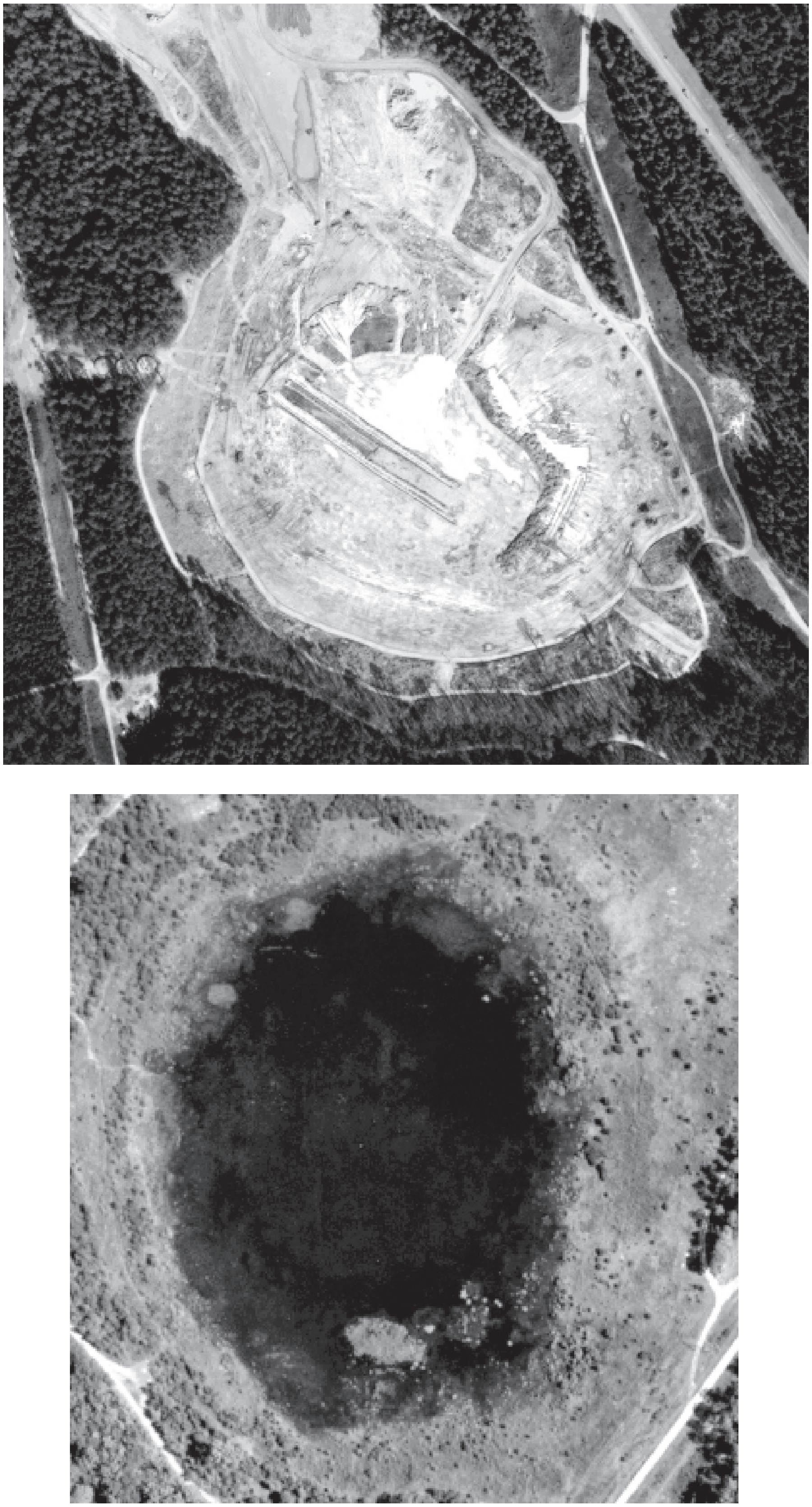

Figure 1. Lost Lake during and after restoration

Table 1. Amphibian species captured after restoration and during the 1977-1978 study by Bennett.

Species	Before Restoration	After Restoration
Salamanders		
Ambystoma opacum, Marbled salamander	X	X
Ambystoma talpoideum, Mole salamander	X	X
Ambystoma tigrinum, Tiger salamander		X
Eurycea quadridigitata, Dwarf salamander	X	
Notophthalmus viridescens, Eastern newt	X	X
Plethodon glutinosus, Slimy salamander	X	X
Frogs and Toads		
Acris gryllus, Southern cricket frog		X
Bufo quercicus, Oak toad	X	X
Bufo terrestris, Southern toad	X	X
Gastrophryne carolinensis, Eastern narrowmouth toad	X	X
Hyla cinerea, Green treefrog	X	X
Hyla gratiosa, Barking treefrog	X	X
Hyla squirella, Squirrel treefrog	X	X
Hyla versicolor, Common gray treefrog	X	
Pseudacris crucifer, Spring peeper		X
Pseudacris nigrita, Southern chorus frog		X
Pseudacris ornata, Ornate chorus frog		X
Rana catesbeiana, Bullfrog	X	X
Rana clamitans, Green frog	X	X
Rana utricularia, Southern leopard frog	X	X
Scaphiopus holbrooki, Eastern spadefoot toad	X	X

tional opportunities. Under the direction of Dr. John Williams, 28 undergraduate and graduate students at South Carolina State University have performed research projects supported by the DOE program for Historically Black Colleges and Universities. At University of South Carolina-Aiken, in addition to the 9 students participating in our research and a post doctoral sabbatical for Dr. Hugh Hanlin, more than 12 undergraduate independent study projects have been performed involving the recovery of Lost Lake. Currently, Dr. J. Hayes of Paine College is studying the insect populations of the hardwood area near Lost Lake to help develop baselines for terrestrial rapid bioassessment techniques. Additionally, two high school students participating in the NSF Young Scholars Program at the Ruth Patrick Science Education Center and the South Carolina Governor's School for Science and Mathematics have done research projects involving Lost Lake. These undergraduate studies have varied in subject matter. A few examples are a study of the mosquitoes of Lost Lake; a survey of small mammals in the Lost Lake wetlands; water quality of Lost Lake; habitat preferences and movements of amphibians such as bullfrogs, mole salamanders and tiger salamanders; bird communities in the Lost Lake wetlands; and comparisons of algae communities between Lost Lake and undisturbed Carolina bays. In addition, over a dozen tours have been conducted by the senior author for groups from area colleges, schools, clubs, and service groups. The recovery of Lost Lake has been the subject of several newspaper articles and will be part of an upcoming CNBC piece on environmental restoration activities at SRS.

To date there have been more than 6 publications in refereed scientific journals and more than 10 papers presented at symposia or meetings of professional societies. These papers and presentations dealt with either changes in amphibians and reptiles following restoration, changes in vegetation, and using wetlands as ecological laboratories for educating students.

Conclusion

The return of Lost Lake has been an example of promises fulfilled. A project that started as almost an afterthought to a waste site clean-up has become an example of how cooperation and initiative can produce effective results in a timely and cost effective manner. Lost Lake was the first Carolina bay to be restored at SRS, possibly the first anywhere. Lost Lake has exceeded its expectations in that it has provided remarkable educational opportunities throughout the local area for high school and college students as well as professors and research scientists. Lost Lake also provided to the science of wetland restoration and resource management a wealth of information on recovery and conservation of these unique wetland systems and environmental restoration in general. In spite of the fact that there are no current on-going research programs at Lost Lake, this aquatic jewel in the sandhills still offers abundant information about how restored wetlands recover and function over time. It is our sincere hope that we can find the resources in the near future to begin again and further investigate the mysteries of how finding Lost Lake has progressed and to enumerate the benefits it offers to the surrounding landscape and beyond to the entire SRS.

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Biographies

Lynn D. Wike

Lynn D. Wike received a BA in biology from Millersville University in 1973, an MS in Environmental Biology from Eastern Illinois University in 1981, and a Ph.D. in aquatic ecology from the University of Illinois in 1987. His training and experience varies widely from ecology to herpetology, fisheries, small mammal ecology, and wetland ecology. He has taught at the university level and continues as an adjunct at the University of South Carolina-Aiken, teaching general biology, zoology, and ecology. He has worked as a biologist at Three Mile Island, the Illinois Natural History Survey, and as a private consultant. He has been the author or co-author of over 30 publications and presentations and has been at Savannah Rive Site since 1989.

F. Douglas Martin

F. Douglas Martin received a BS in zoology in 1964 from Louisiana State University, an MA in 1967, and a Ph.D. in 1968 in zoology from the University of Texas at Austin and an MS in information science from the University of North Texas in 1991. His primary training is in ecology and vertebrate taxonomy. He has taught at university level, worked as a research biologist, resource manager for state and federal agencies, and has worked as a scientist in private industry. He has more than 30 refereed publications including three books or volumes. He has been at Savannah River Site since 1992.

John Gladden

John Gladden received his Bachelors Degree in biology from the University of Pennsylvania in 1970 and his Ph.D. in biology from Emory University in 1979. His dissertation research addressed the movement of radiocesium in a stream-wetland ecosystem on the Savannah River Site. Following completion of his Ph.D. he worked for Texas Instruments, Inc. developing mathematical models for fisheries and carbon cycling in the Hudson River. Subsequently, he had research and research management positions at SUNY-Stony Brook and the University of Georgia's Savannah River Ecology Laboratory. In 1984 he moved to Du Pont's Savannah River Laboratory conducting limnological and wetlands research and impact assessments. In 1989, he became manager of that unit and is currently manager of the Environmental Analysis Section in the Savannah River Technology Center.

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The Evolution of SRSnet

Andrew J. Johnson

Abstract

Beginning in 1982 the site computer network, currently called SRSnet, has evolved over the past 17 years to support the tremendous increase in computing and information technologies at the Savannah River Site (SRS). The growth of this network coincided with the explosion in the number of personal computers for office, laboratory, and process operations use and with the introduction of electronic mail and other office automation tools for site employees. SRSnet expanded throughout the 1980s to cover all site operations areas over the 300-square-mile SRS reservation and has undergone (and still is undergoing) major upgrades in technologies. SRSnet was one of the first large-scale computer networks in the country and remains one of the largest examples of a local area computer network. Because of the distribution of computers and computer-based processing across the Site, SRSnet is now considered essential to Site operations. Current upgrades to the network are focusing on eliminating single points of failure and upgrading the capacity of the network to transport the continually increasing load generated by Site operations.

The Early Years—Savannah River Laboratory and Laboratory Data Automation

The genesis of SRSnet goes back to 1981. At that time, the Computer Systems Division (CSD) of the Savannah River Laboratory (SRL) conducted a study of the automation needs of the Laboratory researchers. In 1981, general purpose computing at the Savannah River Plant consisted of three components. An International Business Machines (IBM) “mainframe computer” processed all major computations, business-oriented as well as most scientific and engineering calculations. A few dozen “minicomputers”, primarily Digital Equipment Corporation (DEC) PDP systems, were dedicated to laboratory data automation and process data monitoring. A small number of microprocessor-based, or Personal Computer (PC), systems were also in use. Although introduced by this time frame, the now familiar pairing of Intel microprocessor hardware and Microsoft operating system software had not yet become the industry *de facto* standard. Personal computers in individual offices were rare, and most site microprocessors were located in laboratories to collect data and manage laboratory instrumentation.

The state of art of computer networking at the Site at this time consisted of a network of a few hundred “dumb” terminal workstations. (“Dumb” being the industry term for a workstation with no independent processing capability.) These terminals were attached through slow-speed phone lines and modems to the mainframe computer. A much smaller number of equally dumb terminals attached via separate phone lines, and modems to a few of the Site minicomputers were also in use.

As part of recommendations to Laboratory management on how to improve the use of computers to support the role of the laboratory researcher, the CSD study recommended establishing a local area network (LAN) based on Ethernet. Ethernet was a networking technology developed in the mid 1970s at the Xerox Palo Alto Research Facility, and, in 1981, it was just beginning to be available commercially. Ethernet defines a set of hardware and software protocols for establishing a communications "bus", linking distributed computers together to pass information and to share peripheral devices such as printers. The basic data transmission rate of Ethernet is ten million bits of information per second (mbps), a rate that was considered extremely fast at the time. In 1982, SRL management authorized the acquisition of a demonstration-scale local area network and supporting computers. The LAN components were purchased from Ungermann-Bass, Inc., and the resulting SRL network represented one of the earliest commercial implementations of an Ethernet network.

For the initial network, the network medium consisted of a 500-meter long coaxial cable that was installed in or near the ceiling throughout key locations in Building 773-A, the principal SRL facility. Network "taps" and "drop" cables connected microprocessor-based "Network Interface Units (NIU)" to the coaxial cable. Computers and terminals were attached via low speed serial cables (RS-232) to the NIUs, also called terminal servers. The Ethernet protocols made it possible for many devices to share a network, each capable of communicating with any other device on the network.

In 1983 the initial SRL network was expanded with the addition of coaxial cable segments, extending the reach of the network to the main laboratory locations of SRL. At the same time a fiber-optic cable and Ethernet repeaters were installed between SRL and the Site Central Computer Facility (CCF), which was located across from SRL. This was the first Site use of fiber optics in a computer networking application, and it provided the capability for micro and minicomputers attached to the network to send data to a minicomputer attached to the IBM mainframe computer. The minicomputer provided a store-and-forward mechanism between the laboratory computers and the IBM mainframe, which contained databases and computer programs for data storage and analysis. Additions of terminal emulation software for personal computers and a "protocol converter" enabled PCs attached to the LAN to emulate either a minicomputer terminal or a mainframe terminal for the purpose of logging into and using one of these "host" computers. By this point, fewer than 100 SRL employees were using the network. However, the network was demonstrating the three primary uses of any computer network:

- Allowing people to access distributed computing resources
- Supporting data transfers between individual computers on the network
- Allowing computers to share peripherals such as printers

Office Automation and the Expansion to All Site Areas

In 1985, Savannah River Plant (SRP) executive management, as the result of a comprehensive study of the computer and communications needs of the Site, formed the Computer Projects Department (CPD). CPD's mission was to establish and support office automation productivity tools for Site "information workers". At that time, the Site population consisted of over 10,000 employees of which 4000 were judged to be information workers by virtue of their job assignments as managers, supervisors, professional, or clerical employees. CPD acquired the office automation system, "ALL-IN-1", a product of Digital Equipment Corporation, which became the basis for the first introduction of electronic mail to the Site. ALL-IN-1 was a complex piece of software that ran on several DEC VAX minicomputers. It required a terminal connection between a user and the host computer. Therefore, CPD had the collateral mission of extending the original SRL LAN throughout the Site so employees at all 12 of the major operating areas would have connectivity to an ALL-IN-1 computer.

At that time, the size of the Site presented a technical challenge to the network implementers. Ethernet remained the state-of-the-art technology for computing network, but Ethernet protocols limited networks to no more than 2500 meters between devices. Since the distances between SRS areas extend up to several miles, simply extending the Ethernet between areas was not possible. Moreover, the state of data connections over existing phone systems at the Site limited transmission speeds on the order of 10,000 bits per second, which was woefully inadequate for linking area LANs together. Using an existing site cable TV (CATV) plant that connected all Site areas for video surveillance and information dissemination applications solved this problem. Specialized broadband/Ethernet interconnect devices, called bridges, were acquired from a small networking company, Applitek. The Applitek bridges transferred data packets between intra-area Ethernet LANs using a 6-megahertz TV channel over the CATV cable system to provide a 10-million-bit-per-second inter-area data network. Also, by this time Ethernet interconnection "bridges" were commercially available. These allowed the construction of complex Ethernet topologies so that the largest site areas could be interconnected via a single bridged Ethernet.

During 1986 and 1987, Ethernet LANs were implemented in each Site area, using Ethernet repeaters and bridges to extend the LANs within each area. These LANs were interconnected using the Site CATV system and the broadband/Ethernet bridges. At this time, SRSnet became one of the largest (in terms of geographical coverage) "campus" networks in the country. Figure 1 is a highly simplified schematic that shows the state of the network in 1987 after it had become a site-wide computer network using extended Ethernet LANs in each Site area and the CATV cable plant to connect the areas together.

Besides the very large area of Site territory, a major factor that hampered widespread deployment of the site network was the network

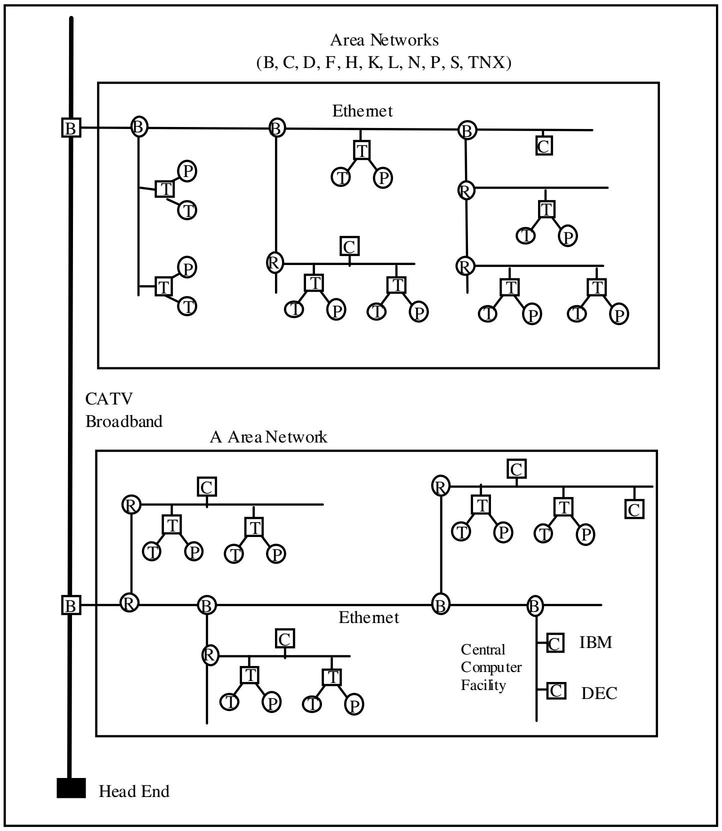

Figure 1. SRSnet - 1987

media itself. Initially, Ethernet required expensive and difficult-to-install coaxial cables. By the mid 1980s, the rapidly growing local area networking industry developed and standardized on a way to use inexpensive wiring consisting of pairs of wires twisted together to improve data communications characteristics. This type of wiring and its installation are common to the telephone industry. A site-structured wiring standard based on the industry standard (10BaseT) was defined, and projects were initiated that eventually resulted in the rewiring of virtually all Site locations so that every office or laboratory has at least one telephone connection and at least two data connection ports. Site rewiring started in 1989 and was not substantially completed until 1996 because of funding limitations.

The first-generation network based on terminal servers providing terminal access to host computers was transformed as the terminal servers were replaced with twisted pair wiring Ethernet "hubs", and terminals were replaced with personal computers containing Ethernet adapters. This meant that user connections were upgraded from a 9600-bits-per-second terminal port to a shared 10-million-bits-per-second Ethernet. Each Ethernet segment typically supported several hundred users. The 10BaseT hubs, Multi Media Access Centers (MMAC) from Cabletron Systems, were joined together by Ethernet coaxial cable or Ethernet extensions over fiber optic links.

By the end of 1989, the Site network had grown to near unmanageable proportions. The network topology consisted of highly complex area networks bridged together over the Site cable TV system. New interconnect devices, called network routers, were introduced to add more reliability and manageability to the Site network. Also by this time the Site telephone system had been upgraded to support high-speed digital data circuits, called T1. A network of routers and T1 circuits replaced the broadband cable system and Applitek bridges. Implementation of the network routers, acquired from Cisco Systems, required that the network be divided into multiple "subnets" with each subnet supporting no more than 255 attached devices. Each subnet was connected to the Site network via a dedicated Ethernet port on a network router. The routers were interconnected to each other via Ethernet segments (within an area) and T1 circuits between areas using a "hub ("A" Area) and spoke" arrangement.

By 1990, a large increase in the Site employee population that occurred after the Site contract transition from the Du Pont Company to the Westinghouse Savannah River Company (WSRC) required the location of several thousand WSRC employees into offsite office buildings. These buildings were located in the nearby cities of Aiken, South Carolina; North Augusta, South Carolina; and Augusta, Georgia. Each of these buildings was wired according to Site standards and equipped with a local area network based on 10BaseT technology. T1 data circuits and Cisco Systems routers were used to connect these facilities to the onsite SRSnet. Figure 2 shows the state of SRSnet after the implementation of routers, subnets, T1 circuits, connection to the Internet, and connections to offsite facilities.

The PC Revolution and WISDOM

As SRSnet was expanding from a few dozen interconnected laboratory computers to several thousand computers and workstations across the entire Site, advances in microprocessor technology made the personal computer ubiquitous with ever increasing capabilities at ever lower costs. This led to the acquisition of large numbers of personal computers at SRS. By the end of the 1980s, thousands of personal computers were attached to SRSnet, yet SRSnet could not really be considered a personal computer local area network. This was because the vast majority of PCs were attached via slow speed serial communications ports (RS-232) and terminal servers to SRSnet. These PCs ran special software that allowed them to emulate a

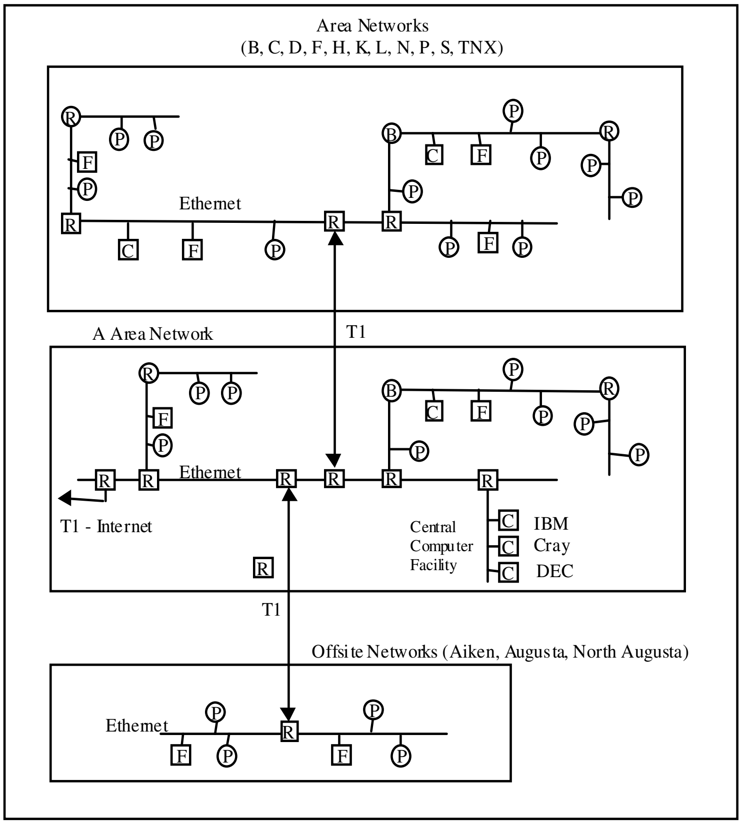

Figure 2. SRSnet - 1992

"dumb" terminal. Computer users employed this terminal emulation software to log onto and use a variety of computers across the Site, including the ALL-IN-1 office automation VAX computers, the Cray scientific supercomputer, and the IBM mainframe.

A true PC LAN consists of personal computers linked at high speeds directly to each other and supporting "servers", computers dedicated to the functions of supplying data storage and printing services for the attached PCs. In 1991, the Site, led by the Information Resource Management Department (IRM), published the first comprehensive computer architecture, to "serve as a unifying plan or blueprint for the implementation of computer-based systems and the computing, communications, and data management infrastructure required to support these systems." The computing architecture document recommended the transformation of SRSnet to a true PC LAN.

In 1992, the WISDOM project became the implementation of this recommendation. WISDOM stood for **W**orkstation **I**ntegration **S**ystem for **D**esktop **O**ffice **M**achines. The WISDOM system consisted of standard personal computers (IBM PS/2 computers with the Microsoft Corporation's Windows operating system and Apple Computer's Macintosh computers) with Ethernet (10BaseT) adapters for high-speed attachment to SRSnet. Local (i.e., nearby) file and print servers provided network file storage and access to network attached printers for all users. Netware, a software product of Novell, Inc., was the basis for providing networked-based file and print services. A standard set of business productivity applications was available for download on demand from the local file server.

The real significance of the WISDOM system was the standardization it brought to Site PCs. Prior to WISDOM, each Site personal computer was set up individually based on the desires of the user or under the direction of some local PC support specialist. This heterogeneous environment made it virtually impossible to deploy modern client/server applications in which the speed and graphical user interface of the personal computer is used cooperatively with the speed and data storage capabilities of larger "server" computers. It also meant that per system PC support costs were very high relative to industry best practices.

In 1996, IRM began to implement standardized PC configurations based on Microsoft's two newest operating systems, Windows 95 and Windows NT. The original WISDOM configurations, based on Microsoft Windows and the Apple Macintosh, were gradually retired. With the introduction of WISDOM and its replacements, the number of personal computers attached to SRSnet grew rapidly from some 4000 in 1991 to over 12,000 by 1996. IRM was able to deploy several client/server applications, and the cost to support individual personal computers rapidly fell to the point that the Site has received industry recognition as "best in class" in its ability to support PCs at a low cost.

The Replacement Telephone System and Fiber Optic Technologies

In 1988, the Department of Energy authorized a major project to completely replace the Site telephone system. This project included replacing all Site telephone switches (PBXs) as well as installing a fiber optic backbone connecting all Site areas together. In addition, the project included fiber optic-based connections to the fiber backbone from all Site permanent facilities (buildings). The "Replacement Telephone System" (RTS) project was completed in 1995. The project was quite an undertaking, consisting of 2 AT&T 5ESS switches with 10 optical remote modules, 18,000 telephone lines, 117 miles of installed copper cable installed, and 184 miles of installed fiber cable.

From a data network perspective, the completion of the RTS project was timely. The rapid growth in the use of information technology, in general, and personal computers, in particular, at the Site had greatly increased the demands for bandwidth (i.e., network capacity) being put on SRSnet. Both the inter-area T1 data circuits and the Ethernet-based trunks in each Site area were being used at near-capacity rates. The new fiber optic facilities opened the door to a newer, faster, and more reliable data networking technology upon which to build the inter-area and intra-area network trunks. This technology is called Fiber Distributed Data Interface (FDDI). FDDI networks are fiber optic based and consisted of rings of systems connected together at transmission speeds of 100 million bits per second. This represents a factor of 10 improvement in bandwidth over Ethernet and a factor of 64 improvement over a T1 circuit.

During the period 1994-1996, a major upgrade of SRSnet was accomplished. Dual inter-area FDDI trunks were used to redundantly interconnect the major site areas. New, much faster routers (Cisco Systems 7000) with FDDI interfaces were the key components of the inter-area network. Each individual area network consisted of several FDDI rings interconnected via a FDDI switch (Digital Equipment Corporation Gigaswitch). The network within each building consisted of several Ethernet (10BaseT) networks connected via a FDDI/Ethernet bridge (Cabletron Systems ESXmim) to the area FDDI ring. Each Ethernet network was limited to no more than 24 connected computers to increase the resulting bandwidth available to any one user. (Previously each Ethernet LAN typically supported 100 to 200 connected computers each). The configurations of the routers were changed to support large subnets, one per SRS

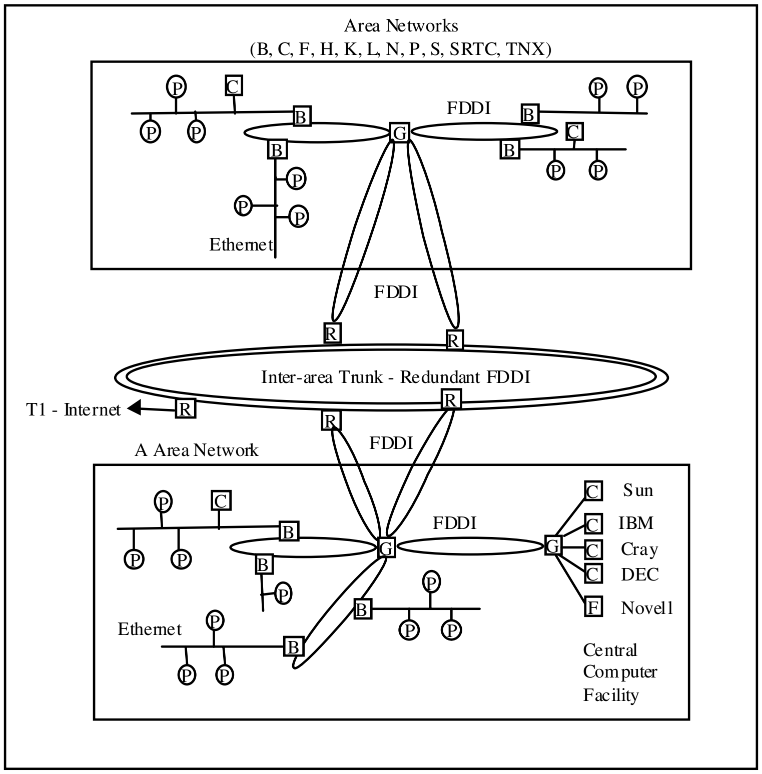

Figure 3. SRSnet - 1997

area. Figure 3 shows the state of SRSnet by the beginning of 1997 after implementing FDDI inter-area trunks, FDDI area networks, large subnets, centralizing the file servers in the Central Computer Facility, and closing the offsite facilities.

This upgrade greatly improved both the capacity and the reliability of SRSnet. The reliability of FDDI technology and the network switches and routers along with the redundant trunk design provided this increase in robustness. Use of the network continued to grow nearly exponentially, and increasingly applications deemed critical to successful Site operations were implemented that depended on the availability and responsiveness of SRSnet services.

The Introduction of the Internet and the World Wide Web

The worldwide computer network now known as the Internet developed out of a Department of Defense research network, ARPAnet, started over 30 years ago. In the early years the Internet demonstrated the utility of electronic mail and the ability to transfer computer files between computers systems across the country (and later the world). In the early 1990s, the development of a new way to present information on a network led to the World Wide Web (WWW, or simply “the Web”) and explosive growth in the size and amount of use of the Internet. Simply put, Web technologies define standard methods of encoding textual and graphical information in a collection of computer files on "Web servers" located across the Internet. These standards enable users on any personal computer attached to the Internet the ability to locate and display for viewing specific information using a PC program called a "browser".

In 1992, SRSnet was connected to the Internet. A secure gateway computer, called a firewall, provided the interface between SRSnet and the Internet. The firewall prevented outside access to SRSnet from the Internet while allowing SRSnet users access to the WWW, external electronic mail addresses, and other Internet services. The initial connection to the Internet was at T1 data rates, but increased usage led to an upgrade to T3 (45 million bits per second) in 1999. The Internet has proven to be indispensable to Site researchers, engineers, and information workers for locating and obtaining information necessary to support Site missions and in communicating with Site overseers, partners, suppliers, and stakeholders.

In 1996, the use of Web technology internally at Savannah River Site was put to use with the **S**avanna**h** **R**iver **I**nformation **N**etwork **E**nvironment (ShRINE). ShRINE is the Savannah River Site's "Intranet" and consists of a system of Web servers containing a wide variety of Site information such as organization charts, telephone directories, newsletters, and electronic versions of common site manuals. A standard browser was installed on every Site personal computer to facilitate employee access to this information and the information found on the WWW.

Since 1996, ShRINE has grown in size and importance to Site operations. Today there are millions of pages of information stored on several ShRINE servers on SRSnet. Over 10,000 users collectively access 2-4 million ShRINE pages in any given week. ShRINE has been extended from the display of static information to include interactive applications in such areas as Material Safety Data Sheets (MSDS), training course registration, personal equipment inventory, and conference room scheduling.

Summary

From humble beginning in the early eighties as a laboratory research tool, SRSnet has grown to become indispensable to Savannah River Site operations. The scope and impact of SRSnet can be seen from the following numbers:

- Area networks - 12
- Buildings served − 600
- FDDI ports − 200
- Ethernet ports − 30,000
- ATM ports - 300
- Fiber cable miles, inter-area - 75
- Fiber cable miles, intra-area - 100
- Connected devices − 15,000
- Number of bytes of data transmitted daily − 8,000,000,000,000 (8 terabytes)

In order to meet the growing volume requirements placed on SRSnet by its users, the next generation SRSnet is currently under development. This is based on new and faster Ethernet protocols supporting transmission speeds up to a billion bits per second.

Acknowledgments

As in any large undertaking, there have been many contributors to the development of SRSnet. I would like to recognize the following individuals (in chronological order) for their major contributions to the birth and growth of the network:

Dr. Randall N. Sims first introduced the concept of local area computer networks to the Site and implemented the first LAN in SRL.

James W. Wade was the Savannah River Plant General Superintendent for Information Systems who sponsored the development of the network during its formative years of 1982-1989.

Frank C. Fortune was responsible for developing office wiring standards that made universal access to SRSnet possible.

James R. Pennington led the SRSnet design and implementation team that took SRSnet from a small demonstration project to total site coverage.

S. Mark Johnson promoted the establishment of an Internet link to the Site and introduced the usage of the TCP/IP network protocols.

David W. Eckert conceived the WISDOM system for supporting personal computers in a distributed LAN environment.

Thomas J. Kennedy designed the major technology upgrade to use fiber optics and FDDI technology.

Philip L. Ames led the efforts to establish ShRINE and use it as a tool to speed the flow of information to site employees.

Henry L. Longley is the current SRSnet engineering lead who was responsible for implementing the 1995-1996 fiber upgrades to SRSnet and who is currently leading the migration of SRSnet to ATM.

Paul N. Sand has been the SRSnet operations lead for the last five years and has been instrumental in implementing past and on-going major network upgrades.

Biography

Andrew (Andy) J. Johnson has a BA degree in mathematics from Rhodes College and an MS in meteorology from the University of Michigan. Following a stint in the U. S. Air Force as a meteorologist and computer programmer responsible for global weather forecast models, Mr. Johnson went to work for the Du Pont Company at the Savannah River Laboratory in the Computer Applications Department in 1977. In his career at Savannah River, Mr. Johnson has been involved in numerous projects, most notably being responsible for the introduction of E-mail across the Site, the early development of SRSnet, the Site computer network, and the formulation of the Site’s first strategic computing architecture. He is currently the Manager for Information Delivery Services in the Information Technology Department of the Westinghouse Savannah River Company.

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Sensor and Detector Technology

C. Wayne Jenkins

Abstract

In the early 1980s, researchers at the then Savannah River Laboratory and later the Savannah River Technology Center began working with fiber optic technologies to use powerful spectroscopic methods for on-line analytical measurements in remote applications. During the past 20 years, these efforts developed fiber optic hardware, sample interfaces, and software integrated with commercially available detectors to form a flexible on-line system for analytical measurements. Methods were developed to analyze solids, liquids, and gases and for physical measurements such as temperature. The system targeted applications in nuclear fuel reprocessing, waste vitrification feed preparation, and weapons surveillance. The advantages were more timely information and reduction in radiation exposure and risks involved with sampling. This technology is in an area of on-going research as more innovative fiber optic probes are developed for measuring new sample types.

The origins of this paper started with developments 20 years ago in fiber optic sensor technology that gained outside recognition with 2 Westinghouse Corporate Signature Awards, an R&D 100 Award, and 16 patents. The scope broadened to include other detector technologies that provided field measurements, particularly the ability to detect and measure radiation. However, the two subjects were too broad for one paper. To do justice to the subject, the author regrettably limited the scope to the original paper.

So, this paper reviews developments in fiber optic technologies by Savannah River Laboratory (SRL)/Savannah River Technology Center (SRTC) researchers that allowed powerful spectroscopic methods, previously confined to the laboratory, to be used for analytical measurements in remote applications. These methods allowed analytical measurements to be made at the point of need rather than relying on time consuming and, in many applications, hazardous sampling followed by laboratory analysis. At the Savannah River Site (SRS), the advantages of these on-line, at-line, or *in situ* analytical methods include reducing personnel radiation exposure and providing timely information to improve control of nuclear materials processing.

Early Attempt at On-line Spectroscopy—Fiber Fluorimetry

In the early 1980s, a vision emerged of automatic control of the SRS reactor fuel and target reprocessing plants, the H Canyon and F Canyon, and the associated product finishing plants, the HB and FB Lines. Computerized distributed control systems were installed, which prompted on-line delivery of analytical information such as chemical compositions as well as traditional pressure, temperature, and flow data. To have significant impact on operations, analytical methods that measured the critical chemical components as accurately and precisely as laboratory instruments would have to be developed, with ruggedness and speed to provide near real-time information. Early attempts to develop on-line analytical systems for the more important constituents, primarily plutonium and uranium, involved investigating fiber fluorimetry. In this technique, laser light was delivered through an optical fiber to a process location. The quantifiable uranium emission was collected by the fiber and returned to a diode array detector. Such a system was demonstrated in the Savannah River Laboratory and included an in-house designed multiplexer, which allowed several sample points to be measured sequentially by cycling the instrument light output and signal input fibers to fiber pairs connected to different sample points. However, the detection limit of 200 ppm in 0.5M nitric acid and 10% uncertainty was insufficient for process control. In addition, the system was not cost-effective to install in the plant since it could only measure uranium.

Though never installed in the plant, the fiber fluorimetry system pioneered three important features needed to practically apply laboratory spectroscopic methods to process or on-line use:

- Using fiber optic cables to remove the instrumentation from the harsh environment (heat, moisture, radiation) of the sample interface
- Using multiplexing to spread the cost of expensive instrumentation over several sample points
- Using the diode array spectrophotometer.

The diode array, which involved no moving parts, simultaneously measured a breadth of wavelengths by projecting the wavelengths of a diffracted light signal onto an array of light-sensitive diodes. The conventional scanning photometric detectors had to rotate a diffracted light signal over a single detector, which made them slow, sensitive to environmental factors, and maintenance problems. The attributes of speed and ruggedness made the diode array spectrometer ideal to use in process areas and for near real-time measurement.

Absorption Spectrophotometry

SRTC scientists recognized that absorption spectrophotometry, in which light is passed through a sample and chemical components are identified and quantified by the extent to which different wavelengths of light are absorbed, could be a powerful analytical tool and more universally applicable than the laser-induced emission-based fluorimetry. However, since many chemicals absorb light at the same wavelength, laboratory applications of absorption spectrophotometry relied upon sample preparation steps to chemically separate the different components before measurement. Since complex feed preparation steps are difficult and time consuming, if not impossible in process usage, applications of spectrophotometry in the process were limited to colorimeters that used one, or possibly up to five, wavelengths to measure specific chemical components in simple chemical systems. If components in a multiple component sample, which most process samples are, had absorptions in the same area of the spectrum, these instruments could not tell them apart. The diode array detector could quickly gather absorption data over a wide range of wavelengths, making it possible to separate components by comparing their absorptions in many wavelengths. However, the volume of data generated by these full spectral measurements and the complicated overlapping absorptions of different chemicals in mixtures made quantifying individual chemical components impractical.

Chemometrics

Also in the early 1980s, SRL became a member of the Center for Process Analytical Chemistry (CPAC) at the University of Washington. CPAC was and still is a member-sponsored research center dedicated to moving analytical chemistry from the laboratory to the shop floor and process environment. CPAC was premier in developing a discipline that became known as chemometrics, the application of what are called multivariate mathematical techniques to complicated data to reduce it to useful information. Chemometrics was applied to sorting complicated spectral information such as that generated in absorption spectrophotometry, to allow individual chemical component concentration measurement. To use chemometrics, one must know what chemicals will be in the mixture he intends to measure and over what range of concentrations each will vary. With this knowledge, a series of calibration solutions is made that bounds all expected chemical components and concentrations. A mathematical model is made from the spectral data obtained by measuring the calibration set. Once the model is made, the mathematics can be used to predict component concentrations from sample spectra. With even the computers available in the 1980s, the analysis was done quickly and within a relatively small instrument package. In addition, since the chemometric model identifies components by comparing the measured spectra with the known contributions of each component to the model, it also recognizes the presence of components that were not included in the model. Although chemometrics cannot identify these unknown constituents, it raises a flag that something unanticipated is in the sample and allows further investigation. The diagnostic capability of a chemometric model-based measurement makes it a robust method.

SRL researchers were the first to practically apply chemometrics. They first wrote in-house software that incorporated the data processing and model building routines necessary to use chemometrics. In cooperation with Laboratories Department personnel in the 772-F process control laboratory, they mated the software to diode array spectrophotometers to use in laboratory measurements of plutonium and uranium. These instruments were much faster and simpler to operate for these process control measurements than the previous nuclear counting and emission-based laboratory methods. Complete measurements could be made in seconds rather than hours with better accuracy and precision. The diode array spectrometer system could take complete spectra every 0.2 seconds. The software could be set up to average spectra over whatever time was desired, usually a few seconds, to remove anomalies and provide statistically sound results. With spectrometer manufacturer hardware updates and software improvements for operator ease, these instruments have been successful for the past 15 years.

Fiber Optic Hardware

Reliable fiber optic hardware had to be developed to extend the use of these instruments outside of the laboratory. SRL researchers developed reliable means of coupling fiber optic cables to instrumentation with minimum loss of light using standard SMA connectors. They also developed techniques to couple the fibers to sample cells using lens assemblies to efficiently collect and focus the incident and signal light. Lens assemblies and fiber optic connectors provided to the Los Alamos National Laboratory (LANL) were used to develop a fiber optic high-acid analyzer that was recognized as one of the year's top 100 technological developments by R&D 100 magazine at the beginning of the 1990s. These fiber optic developments permitted separating the sample cell from the instrument for use in radioactive gloveboxes in the HB-Line analytical facility and in heavily shielded cells such as in Building 772-F. Samples collected in optically clear vials called cuvets could be placed between the fiber optic lenses in a sample holder within the radioactive containment. The spectrophotometer measured the plutonium or uranium concentration outside the containment.

In the next step, researchers incorporated these basic couplings and lenses into standard swageloc fittings to develop leak-tight flow-through sample cells of various sizes and designs that would directly measure a flowing sample or process stream. With the development of these process sample interfaces, absorption spectrophotometry became a viable tool for on-line analytical measurements. These fiber optic on-line systems were first applied in SRL for real-time analytical information on experiments that simulated the operation of the H- and F-Canyon solvent extraction cycles for plutonium and uranium recovery. These systems eliminated the delays of taking samples, sending the samples to the laboratory for analysis, and eliminated the risks of handling radioactive samples. Since the information was essentially available immediately, it could be used to make changes in the experimental parameters such as flow rates and temperatures as the experiment progressed. In other words, the fast analytical information allowed near real-time process control, which was the ultimate goal.

The system described above, consisting of fiber optic flow cells for sample interface, fiber optic cables and couplings, reliable multiplexers, fast and rugged diode array spectrophotometers, and chemometric software, formed a powerful tool with potential for on-line analysis of process solutions in the H and F Canyons. The remaining need was to get the process solutions to the flow cells. By design, the canyon process solutions were contained in vessels and piping behind thick concrete shielding walls. The only access to these solutions was through the individual vessel samplers in the sample aisles in the inhabited part of the canyon, several feet above the process tanks. These samplers were used to obtain samples in small vials, which were transported to the 772-F laboratory for analysis. The samplers operated by pulling a vacuum on a pipe submerged in the vessel while at the same time bubbling air into the submerged end of the pipe. The liquid sample lifted the pipe to the sampler in liquid slugs between air bubbles and collected in the vials. Development work included automating the samplers operation and engineering a fiber optic flow-through sample cell that could be installed on the samplers while still allowing the sample vial to be used in the normal manner. The diode array spectrophotometer speed proved essential for this application because the instrument had to obtain and average several spectra between air bubbles. Otherwise the bubbles interfered with the light transmission through the solution and introduced excessive noise into the signal.

Fiber Optic Uses in Reprocessing Applications

With all the pieces of the on-line spectrophotometer system in place, a system was installed to measure uranium and nitrate in seven tanks of the H-Canyon 2nd Uranium Cycle in 1989. The system operated for a time until most H-Canyon processes shut down after reactor operations were stopped. A similar system was designed and built for the Uranium Solidification Facility that was planned to solidify enriched uranium recovered in the H-Canyon operations for return shipment to the Oak Ridge enrichment facilities. However, Department of Energy policy decisions cancelled operation of the facility before the system was installed. The H-Canyon 2nd Uranium Cycle system is being refurbished at this writing for use with restart of the cycle in December 1999.

After the initial successful demonstration of the system in the 2nd Uranium Cycle, work expanded to using the system to measure other H-Canyon process tanks, primarily those in the "hot canyon". Unlike the 2nd Uranium cycle vessels that handle solutions with the bulk of the radioactive fission products removed, the "hot canyon" vessels handle highly radioactive solutions that pose higher risks for manual sampling and analysis, making on-line analysis more valuable. This goal of a "hot canyon" spectrophotometer system was never realized primarily because of the difficulty of engineering a retrofit fiber optic flow cell for the "hot canyon" samplers, complicated by heavy shielding and remote operating devices. One "hot canyon" sampler was adapted for fiber optics, but the difficulties of retrofitting in high radiation fields with the ever-present risk of high contamination was too costly to continue the program, considering the uncertain future of the "hot canyon" operations.

The Flexible Fiber Optic Spectrophotometer System and Probe Development

We recognized that the fiber optic spectrophotometer system as employed in H Canyon was essentially a modular system for performing spectroscopy over fiber optics and was not limited to absorption measurements in liquids. The components could be exchanged to meet the measurement need. Different light sources, types of fibers, and detectors could be interchanged depending on what measurement was desired and into which area of the electromagnetic spectrum the signal fell. The H-Canyon system used a deuterium light source, 600-micrometer-diameter core silica fibers, and a diode array spectrometer targeted for signal light wavelengths in the visible portion of the spectrum, from 300 to 820 nanometers (nm). For analytes that absorbed light with shorter wavelengths, in the 190-to-300 nm or ultraviolet (UV) region, a xenon arc light source could be used with a slightly extended range diode array spectrometer for a detector. Measurements of solids could also be made by delivering light from a tungsten-halogen source through a probe in contact or close proximity to the solid and collecting the scattered or reflected near infrared light (in the micrometer wavelength range) back through the probe and separate fiber to the detector. Either a conventional scanning spectrometer or an accusto-optical-tunable filter (AOTF) with an indium-gallium arsenide array could serve as a detector.

A special fiber optic probe development by SRTC researchers made it possible to perform fiber optic Raman measurement. In the Raman effect, light at a specific wavelength, such as provided by a laser, when interacting with certain molecules, will be scattered off the molecule at a different wavelength than the incident light. The shift in wavelength from the incident to the scattered light is very specific to the molecule interacting, which makes Raman measurements highly selective and free from interferences. However, only about one in a million photon interactions with the molecule will undergo this effect, which means the Raman signal is weak. One way to enhance the Raman signal is to use a powerful laser. However, large lasers are expensive, take up space, require cooling, and, in general, are not practical for on-line use in the process environment. The other way to enhance the Raman signal is to collect the scattered light more efficiently. The SRTC-designed probe performed this function by combining seven fibers into a single probe. A center fiber was used to deliver the laser light, six surrounding fibers collected the scattered light signal, and returned it to a charge-coupled device (CCD) for detection. The patented SRTC design beveled the collection fiber tips at a specific angle to optimize light collection. It also included replaceable optical filters to remove unwanted light and prevent fluorescence in the signal cable, which could mask the relatively weak Raman signal.

In the 1990s, the flexibility of the spectrophotometer system was exploited by developing more and more probes, such as the fiber optic Raman probe, and sample cells to measure different types of samples. Some of the most important were probes that used a mirror for absorption measurements. These probes combined the light source fiber and signal fiber into the same probe. In the standard absorbence or transmission cell arrangement, the light from the source fiber was passed through the sample solution in a "X" shaped sample flow cell and into the signal fiber. In the mirror probe, the incident light was focused by the end lens through the sample solution, reflected off a polished metal mirror back through the sample solution, lens, and into the signal cable. This arrangement was ideal to use as a dip probe that could be submerged in tanks. Further enhancements of this concept included making the mirror moveable so it could be brought against the lens to take a blank spectra. The blank spectra could then be subtracted from the sample spectra to correct for any film formation on the lens or mirror surfaces. This self-referencing feature added to the already powerful diagnostic capabilities of the system inherent in the software model's ability to detect unanticipated constituents and to use standards and blanks as measurement positions in the multiplexer cycle. Stepper motor-driven mirror probes were also developed that allowed a variety of sample path lengths to be used in situations where the analyte concentration varied over a broad range.

The applications of the fiber optic spectrometer system mentioned thus far have been either measurements in solution or of solids. Cells were also developed for gas-phase absorbence measurements. The in-tank precipitation (ITP) process, which was planned as the means of removing the bulk of the radioactivity from the SRS high-level waste tanks for vitrification in the Defense Waste Processing Facility (DWPF), introduced the possibility that flammable benzene could be evolved from process solutions. To analyze for benzene in waste solutions, a sample cell was developed in which air was bubbled through a liquid sample and cycled through a 10-centimeter tube with fiber optic cables at either end. In this way, absorption measurements of benzene vapor were made in the UV. When the measurements reached steady state, indicating equilibrium of the benzene in the vapor with that in solution, the solution benzene content could be determined. A system of measuring benzene in solution at 30 parts per million was installed in the DWPF laboratory. The method was also incorporated into an on-line system for measuring both benzene and nitrite in the Late Wash Facility, which was to perform the final feed preparation step on ITP material moved from the waste tank farm to DWPF.

Sol Gel Indicator Probes, Physical Measurements, and Other End Effect Probes

The most useful absorption spectroscopic information is found in the visible portion of the spectrum and slightly into the UV. In the long wavelength or infrared region of the spectrum, spectral features for components in solution are masked by the broad and intense absorption of water. In the UV region, the fiber optic cables absorb the light, which reduces the distance the sample interface can be removed from the detector to a few feet as opposed to hundreds or thousands of feet for the visible. Although many chemical compounds absorb light in the visible and near UV portion of the spectrum, many do not. One advantage of the Raman probe is that it allows measuring some chemical species in the visible that do not absorb light in the visible. Another way to measure a chemical species in the visible that has no absorption in the visible (no color) is to react it with an indicator that will produce a product that does absorb in the visible. The use of phenolphthalein indicator to determine pH, which is the concentration of colorless hydrogen ions in solution, by turning red is a simple example. Obviously for on-line use it is highly desirable not to use liquid indicators that require mixing with the sample, waste disposal, and constant addition of the indicator.

SRTC researchers developed a new class of solid materials called sol gel indicators that react with specific chemical species to produce a visible absorbance without the complications of liquid reagents. A sol gel can be thought of as a type of glass formed at low temperature. Since the sol gel does not have to be heated to high temperatures to form its glass properties, it is possible to incorporate organic indicator molecules (most indicators are organic) into them that would be destroyed by the heat of normal glass making. Although the sol gel is a solid, the pore size of the glass matrix can be tailored by manipulating the formation parameters. The indicator molecule is trapped in the gel but the desired species to be measured can still flow through the pores to reach and react with the indicator. The sol gel indicator concept is ideal to use with the fiber optic system since it is a light transmitting solid that can be coated on the mirrored probe lens for submerging in process tanks. Producing a useable sol gel indicator is dependent first on finding an indicator that will react with the analyte of interest. Then the pore size of the gel must be carefully controlled to allow the analyte to reach and react with the trapped indicator (and to dissipate when the concentration of the analyte decreases in the solution) quickly without the indicator leaching out. SRTC researchers demonstrated sol gel indicators for measuring pH, chromium, and uranium.

Just as chemical components can be measured by the changes they cause in light spectra, a physical phenomenon that causes a change in light spectra can also be measured by fiber optic spectrophotometry. While working on one application for spectrophotometric measurement, researchers noticed a cyclical change in the collected spectra from day to day in a long-term test. They ultimately correlated this change with the change in temperature of the fiber optic cables from day to night. This observation led to developing a fiber optic temperature probe based on the temperature-induced change in absorbence spectra of a neodymium doped glass bead at the end of a fiber optic cable. This simple sensor won an award as one of the top 100 technological developments of 1995 as determined by R&D 100 magazine. This sensor can measure temperature over the range of -200°C to 600°C (limited by the softening point of the glass) with accuracy of ± 0.5°C, resolution of 0.1°C, and 0.2°C repeatability. The outstanding feature of the sensor is that it is non-electrical and non-metallic, which means it can be used in applications where electrical or radio frequency interference is a problem or in explosive atmospheres where electrical connections are not desired. A multiplexed group of these sensors was used to measure soil temperature during a SRS experiment in which contaminated soil was being remediated by radio frequency heating.

In 1999, SRTC researchers developed a fiber optic temperature sensor based on a different concept. This sensor measures the change in absorbance of a zinc selenide crystal. Unlike the neodymium sensor, which basically exhibits gradual changes in the absorbance intensity (or more accurately light transmission intensity) over a range of wavelengths, the zinc selenide crystal exhibits a sharp increase in light transmission that occurs over a narrow wavelength band. The band of wavelengths at which the step increase in light transmission occurs changes reproducibly with temperature. The zinc selenide sensor has two primary advantages over the neodymium sensor. First, the sharp change in light transmission is easier to detect than the more subtle changes in the neodymium absorbence and is subsequently less dependent on the quality of the light source, spectrometer, and power of the chemometric model for quantitation. Secondly, since the zinc selenide transmission occurs over a small portion of the visible spectrum, it leaves the remainder of the spectrum available for use by the spectrometer for other measurements. Thus the zinc selenide crystal can be incorporated as one layer of a multiple component sensor in which temperature could be measured along with some other quantity by a single fiber optic probe.

Just as the coating of the fiber optic lens with a sol gel indicator or the use of a crystal or doped glass opens the door to spectrophotometric measurement of quantities that cannot be measured by light interaction directly, the use of active films on the fiber optic probe surface is proving to be a fruitful field of research. In an effort funded by the nuclear weapons complex desire to provide surveillance of warhead components being left for longer durations in the field, SRTC researchers recently developed a fiber optic hydrogen sensor. Fiber optics are ideal for measuring this type because of the lack of a spark source in a potentially explosive atmosphere. This sensor is based on light reflected off a palladium film coated on a fiber optic probe tip. The reflected light spectra change in a manner correlated with the concentration of hydrogen in contact with the palladium. The sensor as demonstrated to date has a lower detection limit for hydrogen of 0.01% in air, reacts in 90 seconds, and returns to baseline in 90 seconds when the hydrogen is removed. At the time of this writing, researchers were still attempting to stabilize the long-term response and repeatability of the sensor at the 0.01% hydrogen level.

To adapt the hydrogen sensor to Site needs, work is underway to find a suitable coating to protect the palladium from the harsh chemical environments of the vapor spaces above the H- and F-Canyon process vessels. A long-standing problem exists of monitoring hydrogen generated in the fuel and target reprocessing plants. Today, these plants are dedicated to stabilizing existing nuclear materials. Operators need assurance that dangerous levels of hydrogen are not accumulating. Air purges are used to ensure hydrogen does not accumulate. However, because of the aggressive nature of gaseous nitrogen compounds generated from the nitric-acid-based processes and the complexity of existing gas analyzers, to date there has been no practical way to continually demonstrate by measurement that the hydrogen is maintained at acceptable levels. The simple fiber optic hydrogen sensor has that capability since stability at the 0.1% hydrogen in air sensitivity level required for the application should be easily attainable compared to the 0.01% target for the weapons application. The present challenge is to protect the palladium from chemical attack.

Another development that SRTC researchers produced for the same weapons application is an extremely sensitive, all fiber optic moisture sensor. This sensor is based on an emerging technology, surface plasmon resonance (SPR) spectroscopy. Under certain conditions, light energy can be coupled (a resonance can be established) into a stack of ultra-thin metal/dielectric layers. The molecule adsorption onto the exposed dielectric film can cause dramatic shifts in the conditions necessary to establish resonance. The resonance may then be optically monitored as reflectivity change, wavelength of a specific feature, or a reflectivity minimum as a function of the incident light angle, and correlated to the degree of absorption of the analyte on the surface. The SRTC moisture sensor uses silica and gold layers in a proprietary arrangement to measure water absorption onto the surface. The sensor can detect parts per million water vapor from 0 to >100% (condensation onset) relative humidity. This sensor has fast response, high sensitivity, low noise, all optical, and intrinsically safe. Potential SRS applications include plutonium metal processing where moisture is a fire hazard and tritium processing where water can form tritium oxide, an extreme health hazard, since the oxide penetrates contamination protection clothing (plastic suits) much faster than the gas.

The continuing work in fiber optic sensors involves developing new probes useable at the fiber optic cable end to allow light to interact with a sample media, whether liquid, solid, or gas, or to react to some physical phenomenon. The modular fiber optic spectrophotometer system for measuring and interpreting light interaction remains essentially the same. The software mentioned earlier is the heart of the system and responsible for its versatility and longevity. The software is updated to interface with purchased components. The software flexibility measures different models for each measurement point such that one point represents a temperature, another a chemical concentration in a liquid, while still another measures hydrogen in air. The software integrates measurement models from calibration information, operates the on-line instrument to make the unknown measurements, and has alarm/control outputs to interface with a process control system. Several commercial software packages today do some of the necessary functions such as the chemometric modeling and are being incorporated into the SRTC system as appropriate. However, none has the flexibility and capability to interface with the detectors and the full range of functions that the SRTC software does. The basic SRTC system remains a powerful platform for continuing sensor developments in the future.

Biography

Wayne Jenkins graduated from the Georgia Institute of Technology in 1969 with a Bachelor Degree in chemical engineering. He came to the Savannah River Site in November 1972 after serving in the U.S. Army and working for the Aluminum Company of America as a process engineer in aluminum smelter electrode manufacturing. He began his SRS career as an engineer in the Separations Technology Waste Management organization with responsibilities in radioactive waste tank farm operations. After over six years in Waste Management and two years as an operations supervisor in the Tritium Facilities, he returned to Separations Technology where he served as an area supervisor for the Multi-Purpose Processing Facility and the FB-Line and then as technical supervisor for the F Canyon and Outside Facilities. He joined the Savannah River Laboratory/Savannah River Technology Center in 1988 where he has since managed the Sensors and Analyzer Technology Group in the Analytical Development Section.

Robotics Applications at the Savannah River Site

Clyde R. Ward, W. Ivan Lewis, Robert F. Fogle, Paul S. Hebert, Phillip J. French, and Frank M. Heckendorn

Abstract

Robots were first developed in the early 1960s with large, hydraulic-powered units and unique, proprietary, control systems. Electric robots controlled by personal computers were introduced in the mid 1970s. By the mid 1980s, robotics technology matured to where nuclear applications could be considered. Personnel at the Savannah River Site (SRS) recognized this opportunity and have applied robotics to many different tasks at SRS since the mid 1980s. These applications significantly reduced personnel radiation exposure and accomplished tasks beyond human capability. This paper provides an overview of five robotics technology applications implemented at SRS, including mobile robots, a pipe crawler, special manipulators, and custom-designed tooling.

Introduction

Robots were first developed in the early 1960s for the automotive industry. These robots were large, hydraulic-powered units with unique, proprietary, electro-mechanical control systems programmed to autonomously perform different tasks. Electric robots with personal computer controls were introduced in the mid 1970s, making them more reliable and less expensive, and popular. By the mid 1980s, robotics technology matured to where nuclear applications could be considered. Also, in the mid 1980s, remotely controlled vehicles with manipulators were developed for bomb disposal in response to bomb threats throughout the world. Though these units are not programmable like a true robot and a human controls all motions, they are typically referred to as mobile robots. Savannah River Site personnel have applied robotics technology to different tasks since the mid 1980s.

SRS was one of the first DOE sites to apply robotics in actual nuclear applications. The driving force was and continues to be reducing personnel radiation exposure, the goal for all mobile robot applications at SRS. However, some tasks such as reactor tank inspection and pipe crawling could be not be accomplished by humans, so this technology was imperative in those cases.

In many applications, robotic systems, such as mobile robots, could be integrated with special tooling and sensors. For instance, to remove a junction box in the H Hot Gang Valve Corridor, a bomb disposal robot was modified and a special cutting tool was added to remove this radioactive junction box with minimal exposure to personnel. The Remote Overhead Video Extendable Robot (ROVER) (see Figure 2), used many years to monitor remote operations in the F- and H-Tank Farms, was created by modifying a standard man-lift for remote operation and adding cameras and lights that can be manipulated. This system saved personnel from radiation exposure with each operation.

In other applications, the lack of commercially available equipment suitable for the task forced the design and integration of commercial components into a unique system. All equipment for reactor tank inspection had to fit through the relatively small penetrations in the reactor tank top, requiring custom design and the capability to endure extremely high radiation exposure during operation. In another

Figure 1. Mobile teleoperator used to remove a wall-mounted junction box

application, the tooling designed at SRS to install pour spout inserts was used with a unique manipulator from a nuclear equipment vendor to maintain the Defense Waste Processing Facility (DWPF) melter. These pour spout inserts have extended the life of the melter to obtain years of additional operation. Although commercial pipe crawlers are available, none could climb about 230 feet through pipes 3 feet in diameter, including vertical sections, or cut off a pipe section after arriving at the remote destination. A unique pipe crawler was designed at SRS to carry a plasma torch to cut a pipe section from a canyon exhaust duct, successfully completing the cut with minimal exposure.

Removing a Contaminated Junction Box

In the late 1980s, a radioactive liquid leak from the F Canyon contaminated the inside of an obsolete junction box in the adjacent Hot Gang Valve Corridor to a level of 200 R per hour. The contamination radiation level was high enough to prevent repairing and maintaining equipment in that section of the corridor. Since the contamination was inside, the box would have to be removed so that normal operations in the corridor could resume. An estimated total exposure of 8 R to personnel would be required to remove the box manually, even using tools mounted on long poles and with the operators behind temporary shielding.

A bomb disposal robot (see Figure 1) was modified by installing a new upper arm assembly and a tool-mounted camera, and by adding two degrees of dexterity in the wrist required to cut the supports and conduits for the junction box. A hydraulic cutter, designed to extract victims from severely damaged cars, was attached to the wrist in three different orientations to make the cuts. Three remote cameras were set up in the corridor for views from different perspectives to monitor the vehicle and manipulator movement.

The junction box was mounted on the wall 7 feet above the floor and was obstructed by several air lines, pipes, conduits, and junction boxes. The mobile robot design was tested and refined by removing a junction box four times in a cold mockup of that section of the corridor. It was also tested by removing an uncontaminated junction box in that section of the corridor and removing a similar, uncontaminated junction box in a different section of the actual corridor. In June 1987, from a control station in the Hot Gang Valve Corridor about 150 feet away from the contaminated junction box, the modified bomb disposal robot made 13 cuts and removed the contaminated junction box in one week with less than 1 R of total personnel exposure.

Remote Observation of Diversion Pits

Radioactive waste produced at SRS is held in large, double-walled, steel storage tanks in F- and H-Area Tank Farms. Operating the tank farms requires manipulating piping and vessels with an overhead crane within below-ground pump diversion pits. Historically, personnel have been required to peek over the side of the pits and give hand signals to the crane operators, who in turn, replaced equipment or moved flow control jumpers. This direct viewing exposes workers to radiation from the pits. When placed on fixed tripods to perform remote viewing, cameras were quite often incorrectly positioned for the views required for the entire operation. As a result, the Remote Overhead Video Extendable Robot (ROVER) was developed and provides multiple overhead video views a considerable distance above the pits. Each camera and light can be remotely repositioned to effectively view the entire operation. ROVER is based on a commercially available, electric, battery-powered man-lift selected because it can position the cameras at an elevation just above portable shielding, as much as 30 feet beyond the shielding, or up to 45 feet above the pits. ROVER deploys two cameras on booms that extend 20 feet horizontally from each other with left and right views. Each camera can remotely pan, tilt, and zoom and is enclosed in a transparent bubble for outdoor use. A third camera, on its own mast, provides an overall view and can be placed up to 10 feet above the other cameras. Two high-intensity lights were installed on the mast for night operations, and all controls and video signals are multiplexed over three coaxial cables. These cables are routed to a van containing the remote equipment; the control console, video monitors, camera controls, lighting switches, two camera boom controls, a single camera mast control, and the man-lift booms, steering, and locomotion.

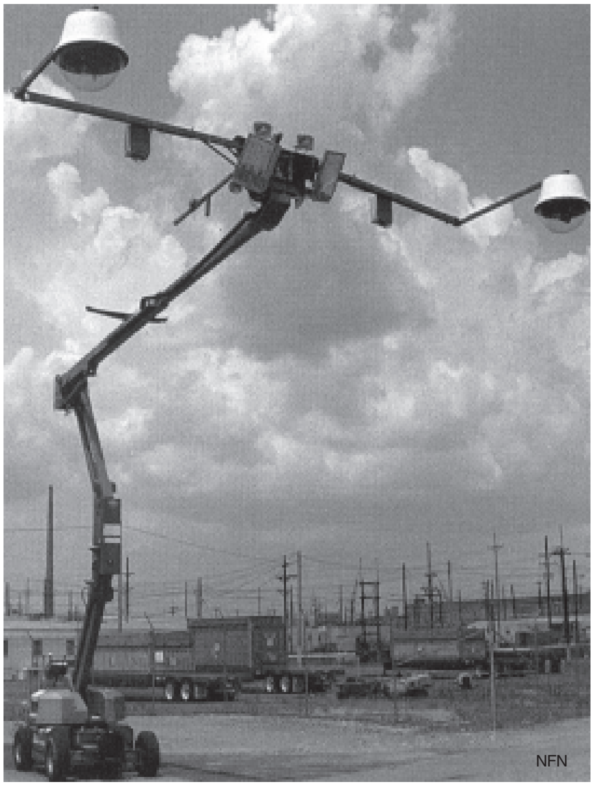

Figure 2. The Remote Overhead Extendable Robot (ROVER) used for

The ROVER mast and cameras can be collapsed to fit within the vehicle footprint for transport to and between F and H Areas. The three camera systems, large distance between cameras, high camera elevations, and repositioning capability have been versatile for remotely viewing the diversion pit operations for many years and reduced or eliminated radiation exposure during diversion pit activities.

Reactor Tank Inspection

In 1989, a program to restart three SRS reactors began, and ultrasonic inspection of the reactor vessel wall welds was a prerequisite to the restart. The reactor vessels were rolled and welded into 1/2 inch thick, 304L stainless steel cylinders 16 feet in diameter. To access the welds, the equipment had to pass through tank top openings 4-3/8 inches in diameter and as much as 21 feet below the tank top, position the sensors with an accuracy of ± 0.030 inches, operate underwater, and endure radiation levels exceeding 1,000 R per hour for days during the inspection operation.

The SRS-designed system (see Figure 3) included an inspection system, the remote inspection robot, three camera positioners, a calibration mast, and a control center designed to be transported from reactor to reactor. The control center was in a 48-foot trailer that housed the robot control, camera control, audiovisual system, and the ultrasonic and eddy current inspection system. A 5-ton crane was designed to be installed, operated, and removed on existing crane rails in the reactor building. Since all components entering the reactor vessel were long with relatively small diameters, strongbacks were designed to transport the components and be placed to the vertical position without damage by a custom-designed erector.

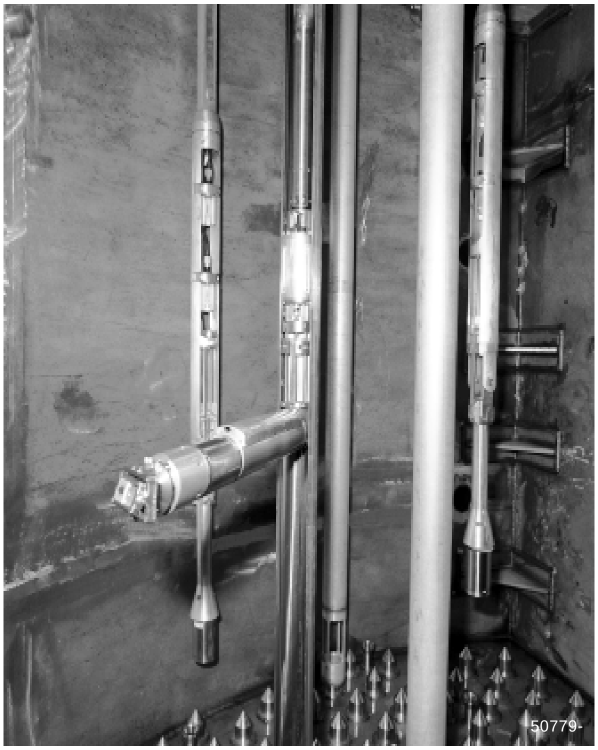

Figure 3. Reactor Tank Inspection Robot used to inspect welds in reactor vessels in

A full-scale mockup of 1/4 of a reactor tank was constructed to develop, test, and qualify the system. The mockup qualified the system in accordance with accepted practices in the nuclear industry and ASME codes. System qualification was achieved using blind samples from an outside agency. During actual operation, a calibration standard on a calibration mast was used before and after data acquisition to assure data quality.

The robotic positioning tool was a five-degree-of-freedom manipulator. The axes included a vertical lift, a rotation about the vertical, an elbow, extension, and a wrist roll. The end effector consisted of two transducers mounted in a gimbal on a spring-loaded, compliant member. A sensor monitored the amount of compliance produced to maintain sensor contact with the wall. Operation of the positioning tool was monitored by three camera and light systems. Each system had a three-degree-of-freedom mast, a radiation-hardened camera, and two lights.

The Reactor Tank Inspection System was successfully deployed in P, K, and L Reactors. It met all performance, cost, and schedule goals.

Pour Spout Insert Installation

Since radioactive start-up, the Defense Waste Processing Facility (DWPF) has experienced glass melter difficulties. The melter, intended to pour vitrified glass mixed with high-level waste long term, was not providing a clean, steady pour. Prolonged glass canister filling caused wicking when the glass stream went over the internal knife-edges of the pour spout. The wicking interfered with glass flow and changing the canisters. These problems were identified during non-radioactive melter testing. Conventional, mechanical master/slave manipulators in the melter cell could not lift the tools and could not reach the melter pour spout. To correct this situation, a custom-designed, electric manipulator with robotic capabilities was installed in existing through-the-wall openings in the melt cell before radioactive start-up. Cleaning tools, a high-temperature inspection device, a removable pour spout insert, and installation and removal tools were developed to use with the electric manipulator to modify the pour spout and reduce wicking caused by pour stream deflections.

SRS personnel designed a high-temperature inspection device to be used by the electric manipulator to inspect the pour spout interior at temperatures beyond 1050 degrees Celsius. Inspection of this device revealed that the upper knife edge that detached the pour stream from the spout was severely corroded. SRS personnel designed and tested a mockup of the pour spout insert at the DWPF operating temperature. The insert provided a new knife edge, closer to the canister opening, for an acceptable pour stream without wicking. The insert was constructed from Inconel 690 to match the pour spout material. The robot uses the insertion tool to place the insert into position and to slide a slotted ring with three horizontal pins up a tapered surface to gain initial contact of the pins against the pour spout. After contact, the thermal expansion drives the pins into the wall, locking the assembly in place. These pins can be pulled, and the insert can be removed after the insert knife edge erodes to the point where a new insert is needed. A chipping tool and scraping tools were designed and used to clean the entire pour spout area between removing and installing inserts.

These pour spout inserts and the tools have been combined with other improvements to increase production and extend the melter life. The increased production exceeded goals set by the Department of Energy and resulted in significant award fees for Westinghouse Savannah River Company. The melter extended life saved millions of dollars associated with changing to a new melter.

Elbow Cutting Pipe Crawler

Because of safety and environmental concerns, an elbow section of pipeline that carried exhaust from F Canyon had to be removed. The elbow was in an underground concrete tunnel that led to a sand filter. By removing the elbow, the air would be redirected from the pipeline's stack to the sand filter. The pipeline was 36 inches in diameter and made of 1/4-inch-thick stainless steel. The section to be removed was 265 feet from the pipeline's entrance. Because part of the building's ventilation system would be affected during the elbow removal process, work had to be timed to prevent a radiological impact on the facility.

A number of metal-cutting technologies were investigated, but plasma arc cutting (PAC) was chosen because of cost, no requirement to introducing liquid, and ease of remote operation.

An internal pipe crawling system was developed to transport the torch through the pipe because the elbow could not be removed externally. The pipe crawler moved in an inchworm motion using pneumatic cylinders. Before this task, SRTC personnel had built units that could operate in pipes up to 12 inches, so this task required a significant scale-up of this technology. A suspension system was added for flexibility to negotiate the elbows and "gooseneck" sections of the pipeline. The resulting crawler was nearly 7 feet long and weighed nearly 125 pounds (see Figure 4).

Six miniature, low-light level, CCD cameras were installed on the crawler to help navigate and locate the elbow section. The plasma arc cutting torch, three cameras, and two lights were mounted to a powered rotator on the front of the crawler. Springs were added to improve compliance in the deployment tool and to compensate for pipe wall irregularities such as weld seams. A tether bundle 300 feet long extended from the rear of the crawler to the control console. The bundle included tubing for the pneumatic cylinders, a torch cable, video coaxial cables, a rotator power cable, a crawler control cable, and cable for emergency retrieval.

After two successful elbow removals in a mockup, the system was moved to a radiological hut that was set up at the pipeline's entrance. The travel time to the elbow section was 2.5 hours. With the aid of the on-board cameras, the first cut was located and made. The crawler was then driven backwards and made the second cut, which successfully dropped the elbow to the bottom of the air tunnel. An insignificant dose was received during the task even though rates of up to 1 R per hour were detected by sensors on the crawler.

Biography

Clyde R. Ward graduated from Tufts University with a Bachelor of Science degree in mechanical engineering in 1996. After working for Du Pont Company in 1966, Mr. Ward transferred to the Savannah River Plant in 1977. Currently, Mr. Ward works in the remote specialty equipment in the Savannah River Technology Center, where he has developed robots, teleoperators, and mobile teleoperators for the Robotics Group.

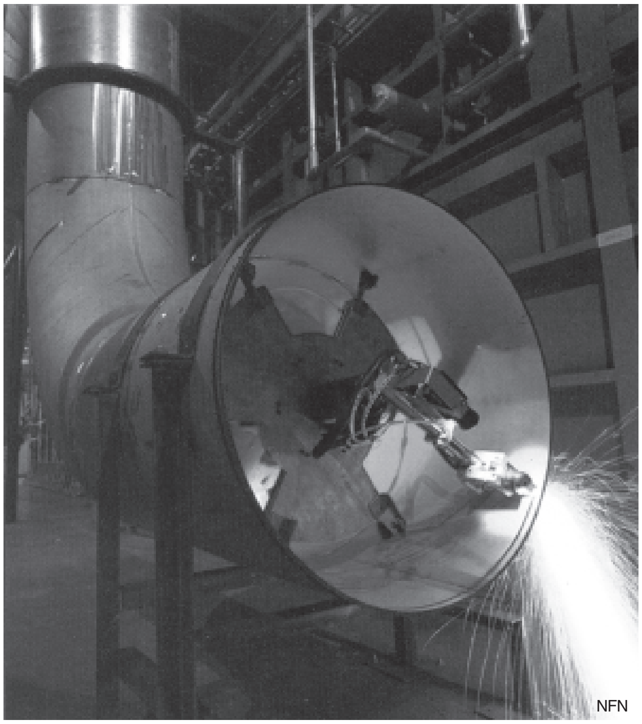

Figure 4. The Elbow Cutting Pipe Crawler was used to cut piping located 265 feet

Future Direction of Science and Technology at SRS

Susan Wood

Abstract

The future for the Savannah River Site (SRS) is bright as we move into the new millennium. Several new missions are underway or are being planned for SRS that will revitalize the Site and set the future direction of science and technology. SRS has been selected as a key asset and resource to continue to support the Department of Energy's three Stewardship programs:

- Stockpile Stewardship
- Nuclear Materials Stewardship
- Environmental Stewardship

Mission responsibilities are projected for at least 30-40 years, expanding considerably beyond the current canyon and facility capabilities present today. Extensive new facility construction projects are planned for plutonium and tritium processing facilities over the next 10 and possibly 15 years.

The technological challenges of the upcoming missions make this an exciting time to be involved in science and technology at SRS. Research, development, and technology application will provide major contributions to ensuring these new facilities operate with today's requirements for nuclear materials management, security, and safety. The Site will exploit the rapid pace of technology development in commercial industry, but reapply these technologies for the specific environments and remote operations unique to SRS processes. Extensive partnering with National Laboratories for joint research and development and technology transfer to SRS is ongoing and will expand. Each stewardship program will be discussed individually with respect to mission and science and technology needs

Nuclear Weapons Stockpile Stewardship

SRS is the only DOE site that has produced tritium for the stockpile and still performs routine tritium recovery and recycle operations. Field units are refilled with purified gas, and excess capacity is stored for future use. Separations and storage processes, which use solid-state technology, rely on continued research and development to support efficiency enhancement.

Because tritium reservoirs are experiencing longer field life, there is an increased emphasis on data collection and analysis from the reservoir surveillance and life storage programs to ensure material behavior can be predicted. This leads to an ever-increasing need for computational tools to analyze large and complex data sets for trends and changes.

In the coming years, the new Stockpile Missions for SRS are expected to include tritium extraction from rods irradiated in the Tennessee Valley Authority (TVA) reactor (the Conventional Light Water Reactor [CLWR] option for tritium production). In the Tritium Extraction Facility currently being designed with a capital investment of $400 million, there are two areas of major technical challenge—remote operations and tritium extraction. Bundles of long target rods received from TVA will be handled, and the entire process will be sequenced remotely. The extraction process will use geometries and temperatures very different from those used in the previous SRS facility because of different target configuration and composition. Process verification and technology integration are key to operational success.

Another potential new mission for SRS is Pit manufacturing, or the manufacturing of plutonium weapons components for the enduring weapons stockpile. Los Alamos has a small capacity for Pit manufacturing but recent Congressional reports delineate a national need for a higher production capability. DOE has stipulated that a new pit production facility will use the process steps employed at Rocky Flats, thus limiting R&D for new materials processes. However, the environmental, safety, and security requirements of this new facility stipulate next generation glovebox operations and scrap recycle. It is anticipated that DOE will begin in the NEPA process for the large-scale pit manufacturing facility by FY02.

Nuclear Materials Stewardship

SRS continues to stabilize and store materials left from the Cold War era, including support for the cleanup of Pu residues and other nuclear materials (including spent research reactor fuel) from other DOE sites such as Rocky Flats and Hanford. This effort requires ongoing flowsheet development for stabilization and disposition of previously unprocessed materials such as Rocky Flats Sand Slag and Crucible. Expanded storage of Pu necessitates new safeguards and securities technologies for surveillance, inspection, and non-destructive assay (NDA). International Atomic Energy Agency (IAEA) oversight requirements drive the development and implementation of remote monitoring and measurement capabilities with real-time data analysis and long distance communication. This is one area where advanced sensors and measurement systems will make a major contribution, ensuring unique signature analysis of each package, its integrity, and security.

The need to disposition surplus plutonium from pits, metals, and oxides has also provided major opportunities for technology development to support the Plutonium Immobilization Program (PIP) and the Pit Disassembly and Conversion Facility (PDCF). Together with the Mixed Oxide (MOX) Fuel Fabrication Facility, these facilities are scheduled for start-up during the period 2006–2008. The PIP will implement a process to produce stable Pu-containing ceramic "pucks" that are subsequently loaded into canisters to be filled with radioactive glass by the Defense Waste Processing Facility. This is the "can-in-canister" concept.

Significant development has already been accomplished by the PIP but the PDCF has some major challenges, particularly given the variety of pit designs it will be required to handle. In addition to scale-up of the metals conversion process, there are major R&D needs in remoting the disassembly of incoming components. In-plant safeguards and security technologies are essential. Implementing processes to meet safety and radiation control requirements needs detailed modeling for design input. SRS is teamed with LLNL, lead laboratory for PIP and LANL and lead laboratory for Pu conversion, in bringing these missions to reality.

SRS will continue to manage research reactor spent nuclear fuel well into this century. "Melt and Dilute"—undergoing development by SRTC—has been recommended as an alternative technology to aqueous processing. Pilot-scale facility implementation in process in L Area and a new Treatment and Storage Facility (TSF) has been proposed for installation in L Area to make the fuel ready for dry storage and transport to the national high-level waste repository at Yucca Mountain. The final decision on spent fuel is imminent. If "Melt and Dilute" is selected, the development work will proceed toward process optimization following pilot-scale demonstration.

Environmental Stewardship

Overall, this area requires technologies for acceleration of cleanup, closure, decontamination, decommissioning; for reduction of risk and cost; and for long-term stewardship. SRS, with its DWPF vitrification facility, leads the DOE complex in the treatment of high-level waste (HLW). We are also meeting or exceeding schedules for cleanup of groundwater contamination and legacy waste sites. But, major challenges remain.

As of this writing, the alternative HLW salt treatment technology has not yet been selected. In partnership with several National Laboratories, parallel R&D continues on options that include small tank tetra phenyl borate (TPB) precipitation and crystalline silico-titanate ion exchange. Once the final down-select has occurred, process optimization will be required. Current and projected HLW inventories at SRS drive the need for an operational facility by ~ 2008.

In addition, new technologies for removing HLW from tanks and new formulations for tank closure are also needed, together with the ever present requirement for improved cost effectiveness for DWPF. Pursuit of increased waste loading and plant throughput (including enhanced system life) are key approaches to accomplishing this latter objective.

For environmental remediation activities, there is a continuing need to deploy new technologies to reduce cost. There is, however, also a significant new emphasis on monitored and/or accelerated natural attenuation for subsurface contaminant cleanup. Application of soft-computing technology, such as neural networks and genetic algorithm technologies, will expand and allow better integration of multi-dimensional factors for determining improved approaches to management and remediation of waste units. Enhanced understanding of exposure pathways and transfer coefficients is necessary to determine appropriate aquatic and sediment compliance limits. Risk management and impact assessment of contamination on environmental media continue to be important areas of study.

Finally, there is now recognition of the need to develop more cost-effective, reliable technologies for long-term monitoring of waste and waste unit closures. This long-term stewardship responsibility encompasses the need to ensure closure integrity or provide corrective actions, if needed.

Decontamination and decommissioning (D&D) will mostly seek to exploit technologies developed by the commercial marketplace. There will be a continuing need to ensure appropriate application and tailoring of these technologies for SRS needs, particularly as the D&D program transitions to dealing with large-scale facilities. There are also areas of technology that are poised for further research and development, such as bioprocesses for chemical decontamination of surfaces.

Summary

Science and technology are critical to the future of SRS. DOE's three stewardship programs and the new missions they bring will set the future direction of Science and Technology (S&T) at SRS. The S&T demands will continue to be varied, covering many areas of research and disciplines, and range from fundamental research to technology tailoring and application. Our strategy for supplying the most effective S&T will be to partner with National Laboratories, universities, and industry. Effective technology transfer will be critical to our success. SRS will ensure implementation of the best and most cost-effective technology to ensure safe, secure nuclear materials management. SRTC will continue to do what it does best: "We put Science to Work."

Biography

Susan Wood, Vice President and Director
Savannah River Technology Center, WSRC

Education
Victoria University of Manchester
Bachelor of Science, physics, 1969
University of Pittsburgh
Master of Science, metallurgical engineering, 1973
Ph.D., materials engineering, 1976

Experience
In her position as vice president and director of the Savannah River Technology Center (SRTC), Susan Wood brings to the Savannah River Site (SRS) many years of experience in industrial research and development executive management in Westinghouse Electric Corporation. In her present assignment, she is responsible for managing SRTC research and development programs and activities in support of the U.S. Department of Energy and SRS operations. The center is an applied R & D organization active in transfer of technology to American industry, industrial partnerships, university partnerships, and economic development initiatives.

She came to SRS from several years in at Westinghouse Electronic Systems in Baltimore, Maryland.; most recently as manager of the Manufacturing Technology Department. She served as the chief manufacturing engineer representing Electronic Systems at Air Force Systems Command.

She also served as general manager of the Materials Technology Division and in other research management positions at the Westinghouse Science and Technology Center in Pittsburgh, Pennsylvania.

She holds patents in materials engineering and has performed extensive work in advanced analytical techniques with a wide variety of materials, and is the author of more than 30 papers for scientific journals, seminars, reports, and presentations.