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**OAK RIDGE
NATIONAL
LABORATORY**

MARTIN MARIETTA

**Chemical Technology Division
Progress Report
for the Period April 1, 1985,
to December 31, 1986**

OPERATED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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**Chemical Technology Division
Progress Report
for the Period April 1, 1985, to December 31, 1986**

R. G. Wymer, Division Director

ORNL-6343

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Date of Issue—August 1987

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ABSTRACT

This progress report summarizes the research and development efforts conducted in the Chemical Technology Division (Chem Tech) during the period April 1, 1985, through December 31, 1986. The following major areas are covered in the discussion: nuclear and chemical waste management, environmental control technology, basic science and technology, biotechnology research, transuranium-element processing, Nuclear Regulatory Commission programs, radioactive materials production, computer/engineering applications, fission energy, environmental cleanup projects, and various other work activities.

As an appendix, the Administrative Summary presents a comprehensive compilation of publications, oral presentations, awards and recognitions, and patents of Chem Tech staff members during this report period. An organization chart, a staffing level and financial summary, and lists of seminars and Chem Tech consultants for the period are also included to provide additional information.

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1. OVERVIEW

The Chemical Technology Division (Chem Tech), which has ~250 staff members, is one of the largest research and development (R&D) divisions at Oak Ridge National Laboratory (ORNL). The Division has outstanding facilities and personnel capabilities in chemical engineering and chemistry, as well as significant competence in several other engineering and scientific disciplines. It has major roles in a broad range of programs, particularly those for which chemical processing is the principal element. These programs are designed to explore selected aspects of nuclear and nonnuclear energy and to develop advanced technology for national priorities. Research and development activities include basic and applied research and engineering, engineering development and assessments, and the operation of various pilot plants and facilities. Most of the research programs are funded by the U.S. Department of Energy (DOE), although some important projects are carried out for other governmental agencies, and limited support is derived from industrial sources.

The overall, long-range objectives of Chem Tech are: (1) to focus primarily on waste and environmental technologies; (2) to make biotechnology, especially bioprocessing, a major activity; and (3) to retain the capability for handling and processing highly radioactive materials. Steady progress in attaining these objectives was made during the current reporting period.

Chem Tech is deeply involved in nearly all aspects of ORNL's activities to clean up and store existing wastes and to plan future waste operations that will be acceptable to state and federal regulatory bodies. The Waste Management Technology Center (WMTc), managed by Chem Tech personnel, coordinates multisite activities in this area and has a major responsibility for participating with industry in the demonstration of commercially available waste management processes. In addition, Chem Tech is involved in DOE's work on permanent federal waste repositories for commercial

radioactive wastes, providing support in systems integration, project management, waste packaging, and transportation. We are also developing and demonstrating the utility of grouts for waste fixation, and we maintain DOE's national data base on present and projected radioactive wastes in the United States. We have recently initiated a new R&D program on hazardous wastes.

Chem Tech's basic research programs in photo-biochemistry, enzyme kinetics, and fixation of enzymes on substrates are internationally respected, and additional funding is expected from DOE in FY 1988 for the bioengineering sciences. We are participating in a collaborative program with the ORNL Environmental Sciences Division and with the University of Tennessee on in situ biodegradation of organic pollutants in the earth. In addition, we have a promising involvement with the Department of Defense for the application of some of Chem Tech's evolving technology for wastewater treatment and in situ waste treatment.

The Transuranium Processing Plant (TPP) continues to be operated by Chem Tech for the isolation, purification, and packaging of Bk, Cf, Es, and Fm for research use; we have also recently put into operation a new hot cell facility to package ^{24}Cl for DOE's ^{252}Cf Industrial Sales/Loan Program. Several initiatives are being pursued to provide additional hot cell programs that will ensure the continued operation of the TPP. We are currently conducting hot cell programs for the Nuclear Regulatory Commission (NRC) on the transport and deposition of fission products from failed nuclear reactor fuel pins.

In addition to the current activities that are supportive of our broad divisional objectives, Chem Tech has active and productive basic research programs in chemistry and chemical engineering. These provide fundamental information and new ideas to the entire scientific community and act as a stimulus for our more applied work.

2. NUCLEAR AND CHEMICAL WASTE MANAGEMENT

During this report period, Chem Tech continued to expand its activities in waste management, undertaking two major new initiatives: (1) the Waste Management Technology Center (WMTC), which serves as the technology demonstration arm of the Oak Ridge Model (a multi-institutional approach to solving waste management problems); and (2) the Hazardous Waste Technology Program, which provides the focal point for ORNL research and development activities concerning hazardous and mixed wastes. Our work for the DOE Office of Civilian Radioactive Waste Management (DOE/OCRWM) continued by conducting additional studies of shales for radioactive waste repository purposes and undertaking analyses and evaluations concerning spent fuel transportation. Simultaneously, Chem Tech was expanding its DOE/OCRWM activities by performing work related to system integration and becoming the lead support contractor for DOE/OCRWM transportation operations. Chem Tech assumed lead responsibility for ORNL tasks designed to provide direction and technology for the management of low-level wastes (LLWs), primarily those generated by DOE. Efforts continue in previously existing areas representing stable waste management projects, such as updating and maintaining the Integrated (waste management) Data Base (IDB) and studying waste immobilization technologies. We also conducted a variety of small, nationally significant waste projects, addressing subjects such as the Three Mile Island Unit 2 reactor cleanup, transuranic wastes, and radioactive waste classification.

2.1 WASTE MANAGEMENT TECHNOLOGY CENTER

During FY 1986, Martin Marietta Energy Systems, Inc., and DOE/ORO established a new Waste Management Technology Center (WMTC), which serves as the technology component of a comprehensive DOE/ORO waste management

coordination effort referred to as the "Oak Ridge Model." The responsibility for the development and management of the WMTC has been assigned to Chem Tech.

The WMTC provides special support in a centralized manner to various waste management organizations in the DOE/ORO-managed complex. The charter of the WMTC encompasses treatment, storage, and disposal technologies relevant to the management of transuranic, low-level, hazardous, and mixed wastes. The WMTC also provides technical support for the development and analysis of technical information supporting remedial actions and compliance strategies by DOE/ORO contractors, including Energy Systems. The expertise available in several of the research and development organizations within Energy Systems is being used in planning and evaluating technology demonstrations and in evaluating the associated implications for the protection of human health and the environment and compliance with regulations. These planning and evaluation exercises are being supplemented by a series of technical workshops that will be conducted by the WMTC to assist in evaluating issues and in assessing potentially useful technologies within a broad context.

The WMTC is emphasizing a technology demonstration program based on cost sharing, which ensures that the expertise of the private sector is efficiently employed while providing answers to current waste management problems on DOE/ORO sites. The facilities managed by DOE/ORO are being used as a laboratory for the demonstration of waste management technologies using actual DOE wastes. It is intended that demonstrations be conducted on scales that will provide credible assessments of cost and performance for full-scale systems and be adequate to determine regulatory compliance. This technology demonstration program provides centralized coordination and quality assurance for the procurement, siting, permitting, and performance evaluation

aspects of technology demonstration projects. Demonstration projects include active participation by state and federal regulatory agencies. The WMTC encourages technology development and application by providing a low-risk demonstration environment through both the partial indemnification of subcontractors and the protection of patent rights associated with the demonstrations on DOE/ORO sites.

The WMTC currently has technical management responsibility for two major programs encompassing all ORO sites and is actively pursuing several related initiatives within the DOE/ORO complex.

The first major program managed by the WMTC is the Low-Level Waste Disposal Development and Demonstration (LLWDDD) Program, which has responsibility for (1) defining and demonstrating improved waste management technologies for low-level solid wastes generated within the ORO complex; (2) siting new disposal facilities to be used by plants on the Oak Ridge Reservation; (3) coordinating the preparation of an Environmental Impact Statement for new disposal facilities on the Oak Ridge Reservation; and (4) designing and constructing large-scale disposal demonstration units within the new facilities. Several LLWDDD demonstration projects were initiated during FY 1986 and FY 1987. These include above-grade, earth-mounded disposal systems based on information exchanges with the French using their experience with similar technology, the evaluation of various waste packaging materials and technologies, and the assessment of mechanical volume reduction technologies based on supercompaction processes. All of these projects have involved participants from the private sector, through competitive procurement processes, with emphasis on the selection of innovative mobile technologies for waste treatment. Scheduled demonstrations of waste immobilization and advanced waste-packaging technologies are being advertised and will be initiated by the end of FY 1987. Demonstrations of diverse treatment, storage, and disposal technologies will continue into the 1990s.

The second major program managed by the WMTC concerns the demonstration of hazardous waste management technologies on DOE/ORO sites. Demonstration projects of this kind are awarded to DOE field offices by DOE's National Hazardous Waste Remedial Action Program

(HAZWRAP) and supported by the sites themselves. The WMTC identifies candidate projects for DOE/ORO, assists in presenting these candidate projects to HAZWRAP for competitive evaluation, and works with DOE/ORO sites to manage any HAZWRAP demonstration projects awarded to DOE/ORO. One such HAZWRAP demonstration project, which was initiated during this report period and will continue through FY 1988 and FY 1989, involves the demonstration of technologies to detoxify (remove the characteristics of a chemically hazardous material) wastewater treatment sludges routinely generated at the Y-12 Plant. If demonstrations of private-sector technologies are successful, sludges that, as produced, have characteristics of both low-level and chemically hazardous wastes can be treated and managed more readily as LLWs by the LLWDDD Program.

Other HAZWRAP demonstration projects involving the remediation of surface impoundments containing organic-laden sludges are anticipated for FY 1988 and beyond. The private sector will be solicited for technological approaches for remediating selected impoundments without creating an inventory of drummed pond sludges that will require subsequent additional treatment.

In addition to the LLWDDD and HAZWRAP activities, the WMTC is pursuing other initiatives that are expected to expand in FY 1988 and beyond. The first of these efforts is to support DOE/ORO plants in site remediation activities by involving the private sector in demonstrations of available or developing innovative technologies. During FY 1987, the WMTC began planning demonstrations of this kind for the closure of the S-3 Ponds at the Y-12 Plant and for the closure of the old Hydrofracture Ponds at ORNL.

With support from DOE/ORO, the WMTC is also establishing, during FY 1987, a special program to involve staff members and graduate students from Historically Black Colleges and Universities (HBCUs) in waste management activities on the Oak Ridge Reservation. The WMTC will work with waste management organizations at ORNL, ORGDP, and Y-12 and with HBCU coordinators for Energy Systems and DOE/ORO to achieve:

1. definition of potential problems to be addressed,
2. identification of HBCU staff members with matching interest and qualifications,
3. development of contractual arrangements, and

4. assignment of HBCU staff members to technical project monitors within individual waste management organizations.

The WMTC will maintain overall management responsibility for this effort and for participating staff members during their involvement in the program. This activity will be expanded to other DOE/ORO sites as it matures.

2.2 GEOLOGIC REPOSITORY ACTIVITIES

In January 1985, Chem Tech completed a comparative evaluation of five sedimentary rock types other than salt for geologic repository siting purposes. This study was initiated at ORNL in January 1984 by DOE/OCRWM to determine the relative suitability of each rock type as a potential host medium for high-level nuclear waste. The rock types considered were anhydrock, carbonate rocks exclusive of chalk (limestone and dolostone), chalk, sandstone, and shale.

To achieve a basis for comparison, rock evaluation criteria derived from the DOE Siting Guidelines (10 CFR Part 960) were applied to each of the rock types, using a generic data base developed for hydrology, geochemistry, rock characteristics (rock occurrences, thermal response, rock mechanics), human interference (natural resources), and dissolution. Based on both postclosure and preclosure evaluation criteria, shale was selected as the most preferred of the five rock types, sandstone was a distant second, the carbonate rocks and anhydrock ranked third, and chalk ranked a relatively close fourth.¹ On the basis of this evaluation, ORNL recommended that DOE:

1. Investigate further the role of shale as a potential host rock for a geologic repository. Investigation of the other four rock types (sandstone, carbonate rocks, anhydrock, and chalk) should be deferred at this time.
2. Develop a data base that would be adequate to proceed with the identification of a potential repository site in shale.
3. Establish cooperative programs with argillaceous rock repository programs in other countries.
4. Examine the potential role of shale in the efforts by DOE/OCRWM to establish geologic repositories for the disposal of high-level waste and spent fuel.

Pursuant to these objectives, a series of three workshops was organized to provide ORNL with an assessment of the state-of-the-art and associated research and development needs in the fields of geochemistry, thermomechanics, and geohydrology as related to repository siting in shale. The first of these workshops, held in Albuquerque, New Mexico, in June 1985, addressed geochemical effects.² The second workshop treated thermomechanical effects and was held in Denver, Colorado, in July 1985;³ and the third dealt with geohydrological effects and was held in Berkeley, California, in August 1985.⁴

As a result of these workshops, technical investigations, involving both laboratory and theoretical studies, were undertaken to expand the geological data base for shales to accommodate the potential need to screen shale occurrences in the conterminous United States for acceptable repository sites. Geochemical studies are directed toward nuclide sorption of key radionuclides in saline systems, aqueous geochemistry, mineralogy of shales, organic material in sedimentary rocks, and natural analogs. Studies in geohydrology include shale permeability measurements, fracture flow modeling, and hydrological modeling of sedimentary rocks. In addition, information on creep, thermal, and mechanical properties of shales is being obtained to increase the thermomechanical data base.

Plans for conducting a national survey of shales, including objectives, technical approach, and methodology, were formulated in mid-1985, but the DOE decision to delay the site selection process for the second repository precluded completion of this activity. Concurrent with the schedule modification, the Sedimentary Rock Program (ORNL) and the Crystalline Rock Program (Battelle) were integrated into the Repository Technology Program

1. A. G. Croff, T. F. Lomenick, R. S. Lowrie, and S. H. Stow, *Evaluation of Five Sedimentary Rocks Other Than Salt for Geologic Repository Siting*, Vol. 1: Main Report, Appendix A and Appendix B, ORNL-6241/V1; Vol. 2: Appendixes C through E, ORNL-6241/V2; Vol. 3, Appendixes F through K, ORNL-6241/V3, to be published.

2. D. G. Brookins and S. H. Stow, *Proceedings of the First Geochemical Workshop on Shale*, ORNL/TM-9865, December 1986.

3. RE/SPEC Inc., *Proceedings of the First Thermomechanical Workshop for Shale*, ORNL-Sub/85-97343/1, March 1986.

4. Berkeley Hydrotechnique, *Proceedings of the First Hydrology Workshop for Shale*, ORNL/Sub/85-37733/1, March 1986.

(RTP) under the programmatic responsibility of the DOE/Chicago Operations Office.

The principal direction of the RTP is the development of mathematical models and computer programs, in concert with laboratory investigations, to analyze the waste isolation potential of crystalline and sedimentary rocks on a non-site-specific basis. Another desired objective is U.S. involvement in cooperative international projects.

In addition to the ongoing technical investigations discussed earlier, ORNL is exploring cooperative programs with Belgium and/or Italy on the use of their underground workings as test facilities for in situ work that would provide extremely important data on the mechanical, hydrological, and geochemical behavior of clay-rich sedimentary rocks. The Boom Clay in Belgium, in which a disposal facility for high-level radioactive waste is planned, has many characteristics that are similar to those of various clay-rich deposits in the United States.

2.3 TRANSPORTATION PROGRAM

The Transportation Program within Chem Tech significantly expanded in scope during this period. Initiatives undertaken by DOE in the civilian spent fuel transportation program and the expansion of ongoing areas of activity resulted in significant growth in transportation. The DOE/OCRWM assigned to DOE/OR the lead responsibility for the development of an operational transportation system to support its spent fuel transportation needs. In this effort, DOE/OR is relying on the Transportation Operations Support Office, which is managed by Chem Tech, to take the lead and to serve as the program integrator for the myriad of activities which are required. The Transportation Operations Support Office is part of the newly created Transportation Systems Program (TSP) established in collaboration with the ORNL Energy Division.

The analysis of hazardous material routing by Chem Tech personnel continued to support analytical efforts related to the movement of chemical weapons by the military and of spent fuel by the DOE. Analyses and drop tests were conducted on designs for radioactive materials packagings. These performance evaluations were directed at determining the capability of specific packaging designs to comply with the regulatory requirements and to investigate the application of computer codes for

analyzing performance. Support was also provided to the DOE activities directed toward the awarding of contracts for spent fuel transportation cask designs to fulfill DOE/OCRWM cask design requirements.

Transportation Operations Support

The DOE/OCRWM assigned the responsibility for the implementation and management of an operational transportation system to DOE/ORO. This system must be capable of transporting the wastes for which DOE/OCRWM has responsibility, including civilian spent fuel and high-level wastes as well as defense high-level waste. All aspects of transportation must be developed (except for package designs), and operational capabilities for highway, rail, and water-borne transport need to be available. In fulfilling this responsibility, DOE/ORO is relying on the Transportation Systems Application Program, managed by Chem Tech, for providing overall support and assistance. In this capacity, Chem Tech has provided direct technical and management support to DOE/CRO, including the integration of other contractor efforts into a cohesive overall effort.

Management Support. Management support was provided to DOE/ORO in planning for the development and management of the transportation system, including the development of activity networks. These activity networks were then loaded onto the CIPREC project management software and were used to develop higher- and lower-level program depictions. Building on the logic networks, preliminary schedules were developed which assisted in the planning and budgeting for future activities. Management support was provided to DOE/ORO for such activities as the development of a project Work Breakdown Structure (WBS) and the definition of elements within the WBS. Subcontractors were often utilized to contribute their specialized expertise in these areas, and support was provided to DOE/ORO in such diverse areas as the development of a project documentation hierarchy, annotated outlines for selected project documents, and complete development of some project documents.

Support was provided to DOE/ORO in the formulation of an overall strategy for the development of the transportation system. This effort consisted of identifying and planning for initiation of development activities. These activities include the

performance of technical and managerial-oriented studies, as well as the identification and resolution of issues such as technical interfaces. The transportation operations system will interface with numerous other entities, both within and outside of DOE control, and, subsequently, must be a very systems-oriented endeavor. Chem Tech recommended and DOE/ORO adopted a strong systems engineering approach to this program.

One of the first major activities undertaken by DOE/ORO at the suggestion of Chem Tech was to study the management configuration alternatives available for the transportation system. A Management Configuration Study was initiated on the basis of a Statement of Work developed by Chem Tech to explore the feasibility, strengths, and weaknesses of the alternatives. The alternatives will be evaluated on the basis of selected criteria and weights that will be determined through a process which includes the consideration of inputs from the public sector.

In order to ensure smooth management of the technical activities associated with the development of the transportation system, Chem Tech also began formulation of a Systems Engineering Management Plan. This plan will provide the guidelines for the overall technical management organization for the project and will be consistent with both higher- and lower-level documents.

Technical Support. In the fulfillment of its responsibility to support DOE/OR in this endeavor, Chem Tech has utilized a strong systems engineering approach in developing the transportation system. Initial efforts in this regard have been directed at the development of a Systems Requirements Document for transportation operations. This activity resulted in the establishment of an overall framework for technical development activities and the identification of key areas requiring further study. The scope of the document included technical issues as well as those related to interfaces and responsibilities. It is serving as the focal point for establishing priorities in the performance of future work.

Technical studies were initiated to define the functions and costs of the transportation system. The Chem Tech staff provided technical oversight for the initiation and performance of a cost evaluation for a cask maintenance facility. This evaluation considered the costs associated with the construction and operation of a cask maintenance

facility which might be sited independently, located on the same site but in a separate building, or integrated into the design of another OCRWM waste management facility. Several issues were raised in this study that will require further investigation due to their potential impact on the design of the transportation system.

Transportation Analysis

During this period, Chem Tech's work on the transportation analysis program has focused primarily on the shipment of radioactive materials. In the past year, however, increased emphasis has been placed on transportation studies related to the movement of chemical weapons for the U.S. Army Toxic and Hazardous Materials Agency (USATHAMA). These efforts are frequently interdisciplinary, as well as interdivisional. The DOE routing studies being conducted in Chem Tech are often accomplished with contributions from the Computing and Telecommunications Division (C&TD). The transport cask studies are carried out in both C&TD and the Engineering Division, while the transportation legislation effort is being handled in the Biology Division.

Routing Studies. A wide range of routing studies was performed in support of the programs of DOE/OCRWM. Three sets of maps were generated, illustrating potential transportation routes between nuclear reactors and candidate repository sites and/or a Monitored Retrievable Storage (MRS) facility. Different modes of transportation (rail or truck) were identified on the maps. The first two sets of maps represented different spent fuel shipment scenarios; that is, although the routes were similar, the number of cask shipments moving along those routes changed. A series of supporting tables was prepared, showing the number of annual shipments from each reactor site, the assumed mode of transportation, and the number of rail and truck shipments expected to pass through each state annually. It was decided to consolidate this information, and therefore, a third set of maps was prepared. In this case, the number of annual shipments along specific transportation routes was shown on the maps; those moving by rail were readily distinguishable from those moving by highway. This change in format resulted in a more comprehensible product.

In order to reduce the manpower requirement and response time, the mapping process was

automated. A series of special computer programs was written to automatically draw the transportation routes and to calculate and automatically label the number of annual shipments along specific routes. After completing the automation process, it was possible to generate a complete set of graphics (photographs, viewgraphs, and slides) for DOE within 2 weeks of receiving its request.

The barge routing capabilities at ORNL were enhanced. A network of inland and offshore waterways has been a part of the rail routing model (INTERLINE) for several years. This network was extensively checked, and the abilities to plot barge routes and to calculate population statistics for subsequent risk analysis were added. This project brings the barge routing capabilities up to the level of development previously attained by the highway and rail routing models. Approximately 120 rail-barge interchange points were identified and added to the network, which will permit the study of intermodal shipments using these two modes.

Transport Cask Studies. These activities constitute highly complex computer analyses of spent fuel transport casks. The computer codes used in the analyses are developed and implemented by C&TD and the Engineering Division under transportation analysis tasks managed by Chem Tech.

Computer Code Benchmarking. Ongoing work in support of computer code benchmark efforts has been organized in the United States by Sandia National Laboratories (SNL) and internationally by the Organization for Economic Cooperation and Development (OECD). The SNL and OECD have both sought to develop analytic benchmark problems that can be applied to spent fuel transport casks through the use of experimental data and, for problems not amenable to experiment, consensus agreement from independent solutions.

The C&TD at ORNL has represented the United States in the OECD working groups on criticality and shielding computational methods. The criticality working group met three times during this reporting period and has primarily been involved with problems arising from potential transportation accident scenarios. The U.S. solutions to proposed benchmark problems have been performed at ORNL using the KENO V.a. code. The shielding analysis working group held its first meeting in June 1986 and discussed solutions submitted to a three-part benchmark problem involving a simple transport cask model. The U.S. solu-

tions were calculated using the DOT IV, MORSE-SGC, and XSDRNP codes. Due to unacceptable inconsistencies in the results from various countries, the problem was reanalyzed using: (1) a variety of cross-sectional data sets available at ORNL and (2) two additional codes (MCNP and QAD) which were used by some participants from other countries. These new solutions, employing five different codes and several cross-sectional data sets, still indicate that a large amount of inconsistency and uncertainty exists in cask shielding analyses.⁵

In support of SNL's thermal benchmark effort and its role in representing the United States in the OECD thermal working group, C&TD at ORNL has used the newest version of the HEATING code (HEATING6.1) to solve a number of cask-related benchmark problems.^{6,7} Excellent agreement has been obtained in comparison with other comparable codes and one available experiment on an electrically heated fuel assembly.

In the structural analysis area, the Engineering Division at ORNL has provided solutions to a set of cask structural benchmark problems designed by SNL. The solutions were obtained using the HONDO-III code.⁸

Related to the benchmark efforts, three studies have been undertaken to better assess the methods used for adequate analysis of casks. These studies include investigating the effect of interface friction on cask impact,⁹ surveying materials and procedures of impact limiters,⁹ and reviewing available

5. C. V. Parks, B. L. Broadhead, R. W. Roussin, J. C. Gauthey, S. N. Cramer, and B. L. Kirk, "Intercomparison of Cross-Section Libraries Used for Spent Fuel Cask Shielding Analyses," presented at Am. Nucl. Soc. Top. Meet. on Theory and Practices in Radiation Protection and Shielding, Knoxville, TN, April 22-24, 1987; to be published in *Proc. Am. Nucl. Soc. Top. Meet. Theory and Practices in Radiation Protection and Shielding*.

6. C. B. Bryan and K. W. Childs, *Thermal Codes Benchmarking-HEATING6 Results*, K/CSD/TM-57, August 1985.

7. K. W. Childs and C. B. Bryan, *HEATING6 Analysis of International Thermal Benchmark Problem Sets 1 and 2*, ORNL/CSD/TM-239, October 1986.

8. C. R. Hammond and T. G. Carley, *HONDO III Code Benchmarking for the Analysis of Impact Response of Nuclear Material Shipping Casks with Extensions*, ORNL/TM-10317, to be published.

9. C. R. Hammond and T. G. Carley, *A Survey of Materials and Procedures for Design of Impact Limiters in Radioactive Material Transport*, to be published.

methods, codes, and data for cask shielding analyses.¹⁰

Burnup Credit Study. An extensive study is being organized by SNL to investigate the incentives and feasibility of taking credit for the reactivity losses due to fuel burnup in the design of the criticality control features of spent fuel shipping casks. The neutronics portion of the study has been performed at ORNL by the Nuclear Engineering Applications Department of C&TD. This effort involved carrying out a series of burnup and radioactive decay analyses to characterize the spent fuel isotopic compositions. These compositions, in turn, were applied in spent fuel cask analyses to determine the cask capacity incentives resulting from reduced reactivity control requirements. Depending on the reference bases in terms of fresh fuel enrichment and cask design, the incentives can range up to 50% in cask capacity. Thus, the potential savings in operations would warrant further effort to qualify the burnup credit approach. The initial work will be documented in an ORNL report¹¹ and two SNL reports.^{12,13}

Cask Shielding Analysis. As part of a study by SNL of public risk from cask transport of consolidated and unconsolidated spent fuel, both gamma and neutron radiation dose rates from two current generation transport casks were determined for various fuel cooling times.^{14,15} The IF 300 rail cask and NLI 1/2 truck cask were selected for this use. All calculations (cross-section preparation, generation of spent fuel source terms, radiation transport calculations, and dose evaluation) were performed using various modules of the SCALE computational system. Conditions or parameters that affected the dose rates include detector distance from cask, spent fuel cooling time, the setting of fuel or neutron shielding cavities to either wet or dry, the cobalt content of assembly materials, the use of normal fuel assemblies or consolidated canisters, the geometry mesh interval size, and the order of the angular quadrature set.

CAPSIZ Computer Code. A personal computer code was developed to evaluate the effects of spent fuel burnup, shielding materials, fuel age, heat transfer, and other factors on cask capacities.¹⁶ The correlations applied in the CAPSIZE model draw heavily on the results of an earlier study.¹⁷ The user can input information on fuel burnup, cooling time, dose rate at 3 m (10 ft) from the cask centerline, the weight limit on the cask, and spacing between fuel assemblies. The output

identifies the cask capacities, assuming the primary gamma shield is built from steel, lead, or depleted uranium. The shield thicknesses and outer diameter of the cask are also displayed. A variation of the program KWIKDOSE can be used to predict radiation dose levels as a function of fuel burnup, cooling time, and cask design features.

Fuel Shipping Studies. One method that has been proposed to reduce the number of spent fuel shipments across the United States to a final repository site, as well as radiation exposure to the public, is that of repackaging the spent fuel in a more compact way. The repackaging could be accomplished at a central location, such as an MRS facility, with the type of packaging (and/or fuel consolidation) dependent upon the geologic media of the final repository. This study¹⁸ determined the cask capacities for 18 predetermined canister sizes and/or overpacks designed to be shipped to repositories employing salt, tuff, and basalt as their geologic medium by utilizing the Shipping Cask Optimization and Parametric Evaluation (SCOPE) code.¹⁷ The assumed cask weight limits were 100 and 150 tons. The spent fuel was assumed to be 10 years

10. C. V. Parks et al., *Assessment of Shielding Analysis Methods, Codes, and Data for Spent Fuel Transport/Storage Applications*, ORNL/CSD/TM-246, to be published.

11. S. P. Cerne et al., *Enrichment and Isotopic Composition of Spent PWR Fuel as a Function of Initial Enrichment, Burnup, and Cooling Time*, ORNL/CSD/TM-244, in press.

12. T. L. Sanders et al., *Feasibility and Incentives for the Consideration of Spent Fuel Operating Histories in the Criticality Analysis of Spent Fuel Shipping Casks*, SAND87-0151, to be published.

13. T. L. Sanders et al., *Incentives for the Allowance of Burnup Credit in the Design of Spent Nuclear Fuel Shipping Casks*, SAND87-0232C, to be published.

14. C. V. Parks, O. W. Hermann, and J. R. Knight, "Radiation Dose Rates from Consolidated Fuel in Current Generation Shipping Casks," presented at Am. Nucl. Soc. Winter Meet., San Francisco, November 10-14, 1985.

15. C. V. Parks, O. W. Hermann, and J. R. Knight, *Parametric Study of Radiation Dose Rates from Rail and Truck Spent Fuel Transport Casks*, ORNL/CSD/TM-227, August 1985.

16. J. A. Bucholz, *CAPSIZ: A Personal Computer Program and Cross-Section Library for Determining the Size and Capacity of Shipping Casks Subject to Various Proposed Design Specifications*, ORNL/CSD/TM-248, to be published.

17. J. A. Bucholz, *Scoping Design Analysis for Optimized Shipping Casks Containing 1-, 2-, 3-, 5-, 7-, or 10-Year-Old PWR Spent Fuel*, ORNL/CSD/TM-149, January 1983.

18. J. A. Bucholz, *Scoping Study of Casks Shipped from the MRS Facility to Various Repository Sites*, ORNL/CSD/TM-237, August 1986.

old, and the gamma shielding in the casks could be lead, steel, or depleted uranium metal.

The results, showing the number of consolidated PWR fuel assemblies that may be shipped in the various types of casks, are shown in Table 2.1. In general, the larger (150-ton) cask was more efficient than the smaller cask, and overpacks placed in canisters prior to shipment reduced the cask payload significantly.

Drop Tests. A type of Three Mile Island Unit 2 (TMI-2) defueling canister, a "knockout" canister, was subjected to a series of drop tests¹⁹ at ORNL's Drop Test Facility.²⁰ These tests were designed to confirm the structural integrity of internal, fixed neutron poisons and, in turn, support a request for NRC licensing of this type of canister for the shipment of TMI-2 reactor fuel debris to the Idaho National Engineering Laboratory (INEL) for the Core Examination R&D Program.

The canister, which will receive and contain fuel-bearing parts of the TMI-2 core for shipment, is built from a 35.5-cm (14-in.)-OD stainless steel pipe, having a total length of 380 cm (12.5 ft). Neutron poisons, in the form of ceramic pellets, are placed in smaller pipes which are supported by internal plates (Fig. 2.1). A full-scale version of this package was loaded with a mixture of water and lead shot to simulate the fuel to be transported

and was drop tested four times from a height of 9 m (30 ft). Three of the four drops were performed with the canister temperature reduced to $\sim -12^{\circ}\text{C}$ ($\sim 10^{\circ}\text{F}$) before release. This temperature reduction ensured that the mass of simulated fuel was located in the canister so as to impart maximum loads to the internals in each drop orientation. Each of the four drops was made with the canister placed inside a cask simulation vessel (CSV) that had foam impact limiters attached to the point of impact. These limiters were designed to reduce the impact loads realized by the canister to those expected if the canister were being transported in the cask which, in turn, was involved in the 9-m drop required by NRC regulations.

Vertical drops were conducted to test each end of the canister, and two drops were made with the canister in the horizontal position. Accelerometer data, photographic records, and X-ray profiles of

19. W. D. Box, W. S. Aaron, L. B. Shappert, P. C. Childress, G. J. Quinn, and J. V. Smith, *Drop Tests of the Three-Mile Island Knockout Canister*, ORNL-6275, September 1986.

20. L. B. Shappert and W. D. Box, "A Drop Test Facility Available to Private Industry," *PATRAM '83, The International Symposium on Packaging and Transportation of Radioactive Materials*, New Orleans, Louisiana, May 15-20, 1983, CONF-830528, Vol. 1, p. 450.

Table 2.1. Number of consolidated PWR fuel assemblies that may be shipped in 100-ton and 200-ton casks

Repository medium	Type of package	100-ton casks			200-ton casks		
		Fe	Pb	U	Fe	Pb	U
Salt							
12 PWRs/pkg	Overpack and canister	24*	24*	24*	36	36	48
12 PWRs/pkg	Canister only	24*	36	36	60	60	72
Tuff							
6 PWRs/pkg	Overpack and canister	12*	8	18	30	30	42
	Canister only	18	24	24	42	42	48
Basalt							
4 PWRs/pkg	Overpack only	20	20	24	36	36	40
4 PWRs/pkg	Canister only	36	40	48	68	76	84
3 PWRs/pkg	Canister only	36	39	42	63	69	75
2 PWRs/pkg	Canister only	34	38	42	62	68	76
1 PWR/pkg	Canister only	31	34	39	55	61	69

*Uses two casks, each containing one waste package and each weighing <50 tons; alternately, one may use a single rectangular or elliptically shaped cask containing two waste packages.

Fig. 2.1. Knockout Refueling Canister.

the internal assembly were obtained for all phases of the drop tests.

Precise physical measurements of the internal poison rods were taken before the canister was assembled and again when the drops were completed and the canister had been disassembled. The two sets of measurements were then compared to determine the permanent deformation in the poison rods from the forces experienced in the drop tests.

A review of the results from the posttest examination and the observations made after each test in the sequence indicates that no significant deformations occurred in the knockout canister as a consequence of the drop testing.

No simulated fuel debris migrated into the region below the lower support plate, nor were any potential fuel escape paths created.

Measurements made on the containment shell after the drop tests indicated it was not damaged in the tests and remained pressure-tight throughout the test sequence.

OCRWM Cask Acquisition Program. The Department of Energy-Idaho Operations Office

(DOE/ID) released a Request for Proposal (RFP) for the design and licensing of from-reactor casks. A Technical Advisory Committee (TAC) was set up to help the Source Evaluation Board (SEB) evaluate the proposals that were received. L. B. Shapert of ORNL was appointed as a member of that committee. The review process was initiated following the submittal deadline of October 31, 1986 and continued for a period of ~5 months, at which time the information generated by the TAC was presented to the SEB. The SEB will then make the appropriate selections based on DOE's needs and the information presented.

Transportation Legislation. The Legislative Data Base (LDB), developed and operated by ORNL, is an on-line resource which contains current information on federal, state, and local legislation which impacts the transport of radioactive materials. The LDB is supported by DOE/OCRWM.

The data base, which operates on the ORNL IBM mainframe computer system, currently contains 1965 legislative actions which have been introduced, enacted, or denied at federal, state, or local levels of government. The LDB staff use 37 data fields to abstract and index various attributes of the legislative actions. Each action is indexed to reflect: (1) geographical areas and political jurisdictions affected by the action; (2) current status and status history of each action; (3) type of transportation restriction specified (e.g., escort, notify, permit, ban); (4) mode of transportation specified; (5) specific subjects or topics identified in the action; and (6) legislative sponsor. The LDB is updated twice weekly.

Reports are prepared on an annual basis, containing citations of all identified laws in effect at the state and local levels. The last report published, which summarized 670 state and local laws, contained indexes for locale, bill number, title word, sponsor, transport mode, transportation restriction, and keywords.²¹ The report also included an appendix containing eight tables with statistical data for the period 1977 to 1985. These data provide an insight into transportation legislation trends for this period.

USATHAMA Transportation Studies. A report on the cost of shipping M55 rockets by truck, rail,

21. N. P. Knox, L. F. Goins, J. W. Fowler, and P. T. Owen, *Transportation of Radioactive Materials: A Summary of State and Local Legislative Requirements for the Period Ending December 31, 1985*, ORNL/TM-9985, April 1985.

or air was completed and then submitted to USATHAMA for review. Comments were received and factored into the final report,²² which identifies the methods that are available to transport the rockets to disposal sites from three storage depots (the Anniston Army depot, the Lexington-Blue Grass depot, and the Umatilla depot) and the costs associated with handling, inspection, dispatching, and transporting them. The shipping destinations considered were Tooele, Utah; Pine Bluff, Arkansas; and Johnston Island. Shipment of the rockets by air was the most complex of the modes examined. Military airports, located both at and away from the continental U.S. storage depots, were evaluated. Truck convoy is a second alternative that was considered in the transfer of rockets to Tooele, Utah, or Pine Bluff, Arkansas, while rail was the third mode examined.

Because of the large number of scenarios proposed by USATHAMA and the need to generate expected costs for those quickly, it was decided to develop a computer model that utilizes the attributes of the shipments (i.e., the scenarios) as input to generate the costs. The program was written for the IBM-PC so that USATHAMA could generate cost numbers for new scenarios it wishes to study. An operating manual is being prepared for the program.

The computer model was validated by generating the cost of shipping M55 rockets to Tooele; the results were identical to those developed manually and shown in ref. 22.

Utilizing the computer program, a set of new scenarios was run to determine the cost of shipping M55 rockets by air from the stockpile stored at their Lexington-Blue Grass facility to Tooele, Utah, under varying conditions.

It was assumed that all rockets would be transported in the single pallet overpack for rocket transport (SPORT). Airport assumptions included an on-base airport at Lexington-Blue Grass and two different airports at Tooele (an on-base field or off-base field located at Dugway Proving Grounds). Two different scenarios were also described for the truck convoys used to transport rockets between Dugway and Tooele. The first case assumed that the weapons would be transported as they arrive in the plane, without an overpack; the second case assumed all weapons would be overpacked at Dugway for the 50-mile trip to Tooele. The results of this study were summarized and presented to USATHAMA.

A value engineering study aimed at determining the type of overpack that is both economical and capable of protecting the shipment of chemical weapons by rail was initiated. Four different overpack designs were evaluated. Weapon capacities varied among designs, which caused the number of trains, and therefore costs, to change. A base case, utilizing the CAMPACT overpack concept, was included for comparison. A draft report,²³ prepared by H&R Technical Associates, Inc., is currently undergoing review at ORNL.

2.4 WASTE SYSTEMS INTEGRATION PROGRAM

The Nuclear Waste Policy Act of 1982 assigned to the DOE Office of Civilian Radioactive Waste Management (OCRWM) the responsibility for disposal of high-level waste (HLW) and spent fuel. A significant activity in the development of the DOE Federal Waste Management System (FWMS) is integrating the components of the FWMS, including storage, transportation, and disposal. The objective of the DOE Waste Systems Integration Program is to ensure that the components of the FWMS will constitute an efficient and safe system that operates as a unified entity. A major factor in accomplishing this objective involves collection of data and development of capabilities used by all program elements.

In support of this objective, the Chem Tech Division manages the ORNL Waste Systems Data and Development (WSDD) Program, providing DOE/OCRWM information and technology with which to integrate the FWMS. Typically, the WSDD activities in the Chem Tech Division have been focused on three areas: waste characterization and waste treatment, system interface studies, and computer modeling.

Waste Characterization and Waste Treatment

The initial efforts to characterize the wastes that the FWMS would eventually handle were initiated

22. L. B. Shappert, D. S. Joy, J. M. Begovich, and I. G. Harrison, *Transport Assessment and Cost Analysis of the Shipment of M55 Rockets to Possible Disposal Sites*, ORNL/TM-9626, October 1985.

23. H&R Technical Associates, Westinghouse Waste Technology Services Division, and Oak Ridge National Laboratory, *A Value Evaluation of the Chemical Ammunition Transporter*, H&R 292-1, January 1987.

in 1985 under the auspices of the DOE Integrated Data Base Program. A technical report²⁴ providing detailed characterization of light-water reactor (LWR) spent fuel was completed in 1985. In December 1985, the WSDD Program was initiated as an extension of previous efforts. This effort primarily involved collecting very detailed information on LWR spent fuel and HLW and miscellaneous wastes that might also be sent to a repository. This effort included reviewing data bases of DOE's Energy Information Administration (EIA), the Electric Power Research Institute (EPRI), the U.S. Nuclear Regulatory Commission (NRC), and the Nuclear Materials Management and Safeguard System (NMMSS), as well as placing contracts with industry fuel assembly vendors, to obtain this kind of information. These data encompassed LWR spent fuel and assembly hardware, immobilized commercial (West Valley) and defense HLW, other spent fuels, and special waste sources such as activated metals and isotope capsules. A draft report²⁵ incorporating this information was issued, along with three user-oriented personal computer data bases. For the waste treatment data development activity, the waste acceptance criteria were reviewed, the physical and chemical properties of the waste (determined in the characterization effort) were categorized, and a draft report²⁶ was issued. All these reports were issued in 1986. Future efforts in the waste characterization/treatment area involve extending this data development activity into defective LWR fuel and detailing LWR fuel assembly hardware, as well as updating the other data.

System Interface Studies

Interfaces between the nuclear reactor waste generators, the waste transportation system, and the waste repository can have a significant impact on the FWMS performance since a common item, the cask, must be handled by all of them. In an effort to understand and characterize these interfaces, the Facilities Interface Capability Assessment (FICA) subtask of the WSDD was initiated in 1986. The primary emphasis of this subtask during 1986 was the selection of a subcontractor to perform a two-phase study. Nuclear Assurance Corporation was selected in a competitive procurement process, and the Phase I data collection activities were initiated. These activities consist of (1) generating a master list of data parameters to

determine the type of reactor information desired, (2) coordinating with various groups (EPRI, Pacific Northwest Laboratories, Office of Nuclear Waste Isolation, Sandia National Laboratories, Weston, and others) to obtain agreement on information needs, and (3) searching the open literature and licensing documents to obtain the desired information. The Phase I data collection activities will be completed in 1987, after which the Phase 2 activity, verifying the data with utility nuclear power plant on-site visits, will begin.

Modeling

A number of common technical models and computer codes are required by different parts of the FWMS for design and licensing of various facilities. FWMS integration and economic benefits are achieved by using the same computer codes in these different applications. Our modeling task (1) identifies these generic methodologies and capabilities, (2) assesses their current status with respect to FWMS and NRC licensing requirements, (3) identifies required development and quality assurance (QA) work, and (4) performs the required computer code development. During 1986, an assessment of computer codes commonly used throughout the FWMS was initiated. Two draft reports^{10,27} on this assessment were submitted to DOE/HQ for review. These assessments will continue in 1987.

Efforts to validate the ORIGEN2 code were also initiated. ORIGEN2 is the primary code for determining radionuclide inventory, decay, and generation; it provides input into many other computer codes. A draft report entitled *Radiological Characteristics of LWR Spent Fuel: A Literature Survey of Experimental Data*²⁸ was submitted to

24. J. W. Roddy, H. C. Claiborne, R. C. Ashline, P. J. Johnson, and B. T. Rhyne, *Physical and Decay Characteristics of Commercial LWR Spent Fuel*, ORNL/TM-9591/V1&R1, January 1986.

25. K. J. Notz, A. R. Irvine, R. Salmon, and T. D. Welch, *Characteristics of Potential Repository Wastes*, ORNL/TM-10213, to be published.

26. A. R. Irvine, *Predisposal Treatment of Miscellaneous Radioactive Wastes*, ORNL/TM-10214, to be published.

27. R. M. Westfall, H. R. Dyer, and L. M. Petrie, *Assessment of Criticality Computational Software for the DOE Office of Civilian Radioactive Waste Management Applications*, ORNL/CSD/TM-247, to be published.

28. J. W. Roddy and J. C. Mailen, *Radiological Characteristics of LWR Spent Fuel: A Literature Survey of Experimental Data*, ORNL/TM-10105, to be published.

DOE/HQ for review. An ORIGEN2 preliminary QA plan has been written and submitted to the DOE Oak Ridge Operations Office, and plans are being made for experimental hot cell activities to validate ORIGEN2 code predictions.

Waste Systems Data and Development Program Summary

The WSDD Program was initiated to obtain the information necessary to make FWMS decisions and trade-offs and to develop software and hardware common to the FWMS elements. These efforts will continue into the future, with some of the focus shifting to the system integration of these information and technology tasks. The large number of different waste generators, regulatory agencies, and political jurisdictions involved make the systems integration task important to this program.

2.5 HAZARDOUS WASTE TECHNOLOGY PROGRAM

The Hazardous Waste Technology Program (HWTP) is a new ORNL program that was created in 1986. The HWTP is responsible for programmatic activities concerning hazardous and mixed waste technology, including (1) research and development (R&D); (2) analyses, assessments, and evaluations; and (3) technical support not related to demonstrations. The program involves several divisions at ORNL (Chem Tech, Energy, Engineering Technology, Environmental Sciences, and Health and Safety Research) and will be expanded to include other divisions as their particular areas of expertise are needed. In addition to ORNL, the Y-12 Plant is active in the program, and the Oak Ridge Gaseous Diffusion Plant (ORGDP) is interested in becoming involved. Program personnel work with sponsors to define needs in the areas mentioned above, convey the sponsors' needs to the Oak Ridge technical community, and represent Oak Ridge's capabilities to meet these needs. Ongoing work is monitored to ensure that the results are timely, within budget, integrated across organizational boundaries, and of high quality. In carrying out these responsibilities, the program interacts with agencies other than DOE in areas where the skills of Oak Ridge personnel can be effectively applied.

The HWTP interacts closely with the Hazardous Waste Remedial Action Program (HAZWRAP), which is responsible for reviewing the DOE sites for hazardous wastes; deriving planning and summary documentation; sponsoring research, development, and demonstrations to solve DOE problems; and assisting in the remediation of the DOE sites. The HAZWRAP is the DOE representative to a working group comprised of members from the U.S. Department of Defense (DOD), the U.S. Environmental Protection Agency (EPA), and DOE, which coordinates federal agency hazardous waste activities. Some joint projects are conducted through this group in which ORNL is called upon to assist. The HAZWRAP also works closely with the DOD to manage remedial action planning and to funnel technical problems into ORNL. The HWTP serves as the focal point for the R&D conducted by ORNL for the HAZWRAP.

The significant hazardous and mixed waste problems at the DOE-Oak Ridge Operations (DOE-ORO) sites resulted in the establishment of the Oak Ridge Model, which is a corporate means for addressing the technical needs of the cleanup activities. The Waste Management Technology Center (WMTC) is a cornerstone of the Oak Ridge Model. The WMTC, a complementary organization to the HWTP, manages all the demonstrations of private sector technology applicable to the solution of the hazardous and mixed-waste problems for ORO. Research and development projects conducted by the HWTP logically lead to demonstrations by the WMTC, and some demonstrations may require additional development studies before implementation. The full utilization of both these organizations will provide better technical options and can help maintain diverse, high-quality staff to support Oak Ridge programs.

The Central Waste Management Office (CWMO) coordinates the activities of all ORO sites to ensure that hazardous and mixed-waste issues are addressed uniformly. Some R&D on problems generic to ORO installations is coordinated through the CWMO, while the technical management is provided by the HWTP. The technical staff members from all the DOE-ORO sites work collectively through the HWTP, and this corporate approach is assisted by the CWMO.

All the organizations described above are part of the Nuclear and Chemical Waste Program

(NCWP) of ORNL. This organizational structure is addressing the problems of DOE and other agencies in a synergistic manner.

Current Projects

Several sponsors support the R&D and technology studies managed by the HWTP. This section will summarize some of the current work to illustrate the function of the program.

Study of Options for Nuclear Reactor at Wright-Patterson Air Force Base. The Air Force Logistics Command requested ORNL to study a range of remedial actions that could be taken on an entombed reactor and to recommend which action should be taken. The four scenarios under study vary from tightening up current procedures and maintenance practices to a complete demolition of the reactor and building. This substantial effort is being undertaken by a technical team from the Chem Tech, Energy, and Engineering Physics and Mathematics divisions. The Air Force will choose an option in FY 1987, and the option will be developed in FY 1988 by the project team.

Remediation of Contaminated Groundwater at McCord Air Force Base. The Air Force Military Airlift Command has a base in Washington which has underground aquifers contaminated with jet fuel. The Environmental Sciences Division is studying the base hydrology, directing an in-depth site characterization of the base, and investigating pump-and-treat technologies to remove some of the fuel. Chem Tech is determining whether in situ biological destruction techniques can destroy the residual fuel remaining after the pumping. Some laboratory experiments are planned to supplement the review of current technologies being conducted.

Air-Stripping with Emission Control. The Air Force Engineering Services Center has asked Chem Tech to assemble portable unit operations equipment that can be transported to various Air Force bases and used to develop techniques to remove volatiles from the stripping air by catalytic converters, regenerative carbon adsorption beds, or canister carbon adsorption beds. The air-stripper will be either a packed column to evaluate five column packings or a novel centrifugal contactor. Detailed procedure manuals and operating cost data will be collected during this 3-year project.

In Situ Soil Venting. The Air Force Engineering Services Center is sponsoring a technique called "in situ soil venting" which vacuums volatile organics

from porous, unsaturated soils. High-pressure air is injected into wells that surround the vacuum well. Prior studies by the U.S. Army indicate that ~75% of the volatile organics in a plot may be removed by this method. This study, at two Air Force bases as yet undecided will determine proper well spacings, vacuum and pressure settings, off-gas treatment methods, and long-term operating costs. This 3-year project is being conducted by the Environmental Sciences and Chem Tech divisions.

Hazardous Waste Thermal Treatment with Energy Recovery. The Air Force Regional Civil Engineer—Dallas has contracted with the Energy Division of ORNL to conduct a study on the destruction of hazardous waste by thermal treatment, with the heat recovered from this high-temperature process to be used to produce electricity. The study assumes that the waste site also has a municipal waste burner that incinerates garbage to produce electricity.

Waste Minimization via Plastic Abrasive Blasting. The Naval Energy and Environmental Support Activity (NEESA) has requested ORNL assistance in minimizing waste generation by substituting plastic media blasting for methylene chloride paint stripping. ORNL is providing engineering and ventilation studies support in testing a plastic media blasting facility at a Navy base in North Carolina. This project is being conducted by the Engineering Technology Division and the Engineering Organization.

Paramagnetic Separation of Hazardous Wastes. The DOE Defense Program is sponsoring the Engineering Technology Division in developing methodology for the removal of slightly magnetic heavy metals from sludges and mixtures by using magnetic forces on a sloped chute. Good results have been obtained in separating uranium from magnesium slag produced at the Feed Materials Production Center at Fernald, Ohio; and studies on removing chromium from ORGDP pond sludges are under way. This work should lead to a demonstration in FY 1988.

Tennessee Valley Authority Hyperfiltration Studies. ORNL is teaming with the Tennessee Valley Authority (TVA) and Associated Technologies, Inc., to develop and construct a pilot unit using hyperfiltration to remove radioactive waste constituents from wastewater at a nuclear power plant. In this technology a dual-layer membrane of zirconium(IV) oxide-polyacrylic acid is used over a porous, stainless steel support. Clean water is

forced through the membrane, and the contaminants are retained. The volume of radioactive waste is reduced as much as 20-fold by this process, thereby decreasing the load on the radioactive waste evaporator or other treatment process.

Several process parameters were dynamically studied, including rejection of group beta/gamma activity, total suspended solids, total dissolved solids, and boron. The pilot results were encouraging and agreed with values predicted by hyperfiltration models. From this pilot information, the nuclear power plant operator can vary operating conditions to meet specific plant objectives. The skid-mounted hyperfiltration system will be installed at the TVA Watts Bar plant in 1987.

Hazardous Waste Minimization at the Louisville Naval Ordnance Station. NEESA has asked ORNL to study the wastewater treatment systems at the Louisville Naval Ordnance Station (NOS) and to recommend state-of-the-art methods to treat hazardous waste discharges. Wastes from the NOS plating shop presented several problems: (1) chromium contamination in the wastewater, (2) potential organic contamination from paint-stripping operations, (3) inadequate characterization of the effluent wastewater, and (4) large quantities of water to be treated. ORNL has evaluated several methods to treat the problems and has recommended treatment methods to NEESA. System design and implementation are planned for 1987.

Westover Air Force Base Wastewater Pretreatment. The Air Force Reserve asked ORNL to address wastewater treatment at their base in Chicopee, Massachusetts. ORNL conducted on-base studies that revealed measurable concentrations of oil, grease, and paint-stripping solvents. Results of an analysis of water loading on the municipal sewer system and recommendations for improvement were delivered to the Air Force Reserve.

New Initiatives

Several activities under way at the end of the report period will expand and strengthen the HWTP. Some of these activities are described in the following paragraphs.

Y-12 In Situ Biological Destruction of Chlorinated Waste. DOE-ORO and the Y-12 Environmental Group are funding a team of investigators from the University of Tennessee, the Oak Ridge Research Institute, and the Environmental Sciences

and Chem Tech divisions at ORNL to conduct field research on destroying complex chlorinated hydrocarbons, including polychlorinated biphenyl (PCB), at various oil spills at the Y-12 Plant. Levels of various bacterial strains, nutrients, oxygen, and fertilizer will be studied in contained drum lysimeters. Successful development of this technique could lead to a demonstration project.

Y-12 Terrestrial Ecology Studies. The Environmental Sciences Division hopes to conduct two projects sponsored by the EPA at Y-12. Kerr Laboratories is funding a study of the migration of oil from old oil spills, such as those associated with the Z-oil system at Y-12. The National Institute of Environmental and Health Sciences (NIEHS) may fund a team from ORNL, the University of Tennessee, and the University of Illinois for 5 years to study waste uptake by large and small mammals and microorganisms at a controlled waste site. The old oil trenches at Y-12 meet the site criteria and have been proposed to the NIEHS.

Toxic Substances Control Act Incineration of Low-Level Waste at ORGDP. A goal of the DOE Low-Level Waste (LLW) Program is to investigate the fate of LLW and mixed LLW-hazardous wastes in an operating incinerator. The ORGDP Operations and Process Support divisions are teaming with Chem Tech to propose that this work be done at the new Toxic Substances Control Act incinerator at ORGDP. This 3-year project will begin in FY 1988.

DOD Remedial Action Planning Preparation. The Health and Safety Research, Environmental Sciences, and Chem Tech divisions are jointly conducting the site characterization and remedial action planning for Air Force bases as part of the Air Force Installation Remediation Plan. Each plan requires a multi-year effort. R&D needs identified during this planning project will be proposed to the Air Force for additional funding.

DOD Remedial Action Plan Review. The HAZWRAP has requested the Oak Ridge technical divisions to technically review the remedial action plans prepared by outside contractors for completeness and appropriateness before submittal to the respective DOD agency. Engineering reviews will be conducted by the ORGDP Process Support Division and by Chem Tech. Earth sciences reviews will be performed by the Environmental Sciences and the Health and Safety Research divisions.

Private Sector Involvement. Some nonfederal queries have been received requesting ORNL support for hazardous waste R&D; these queries are being investigated. A major electronics company is interested in ORNL expertise in using lasers to develop instrumentation for destruction or concentration of hazardous wastes. A state study group is interested in ORNL grouting and incineration technology for possible application at a regional incinerator facility. These questions are coordinated with the Business Development staff and the affected research staff. Although this work will likely remain at a minor level, these tasks will be carried to completion since the transfer of technology is a goal of ORNL.

Complex Hazardous Waste Technology

The progression from hazardous waste R&D to the application of developed technologies to specific environmental problems involves the cooperative interaction of multiple disciplines and divisions. This cross-cutting of the normal discipline-oriented divisions into project teams is complex and requires more cooperation and integration than do many ORNL tasks. The management of these complex tasks is a prime purpose of the HWTP.

ORNL has requested the DOE Office of Planning to commission ORNL to evaluate the R&D needs for hazardous waste treatment and disposal. This in-depth study, with recognized experts being used as consultants, will require about a year to complete. The document resulting from this study will assist DOE in deciding R&D priority areas and will evaluate the appropriateness of current programs.

The HWTP, although in place for only a short time, is providing a needed focal point for research divisions and sponsors to interact. This program is expected to grow and to become more established during the next report period.

2.6 INTEGRATED DATA BASE PROGRAM

The Integrated Data Base (IDB) is the official DOE data base for spent fuel and radioactive waste inventories and projections through the year 2020. The IDB Program is jointly sponsored by DOE's Office of the Assistant Secretary for Nuclear Energy, Office of the Assistant Secretary

for Defense Programs, and Office of Civilian Radioactive Waste Management. The purposes of the IDB Program are to create and to maintain a reliable baseline of quality data and information to be used for national program planning, decision making, and other management activities. As the official data base for DOE, the IDB is readily available to the DOE community and to DOE-sponsored contractors. Work on maintaining and upgrading the IDB continued during this report period.

The IDB Program provides access to information on spent fuel and radioactive waste inventories and characteristics, including volume and/or mass, age, radioactivity, heat generation, chemical and physical properties, location, packaging, and nuclide composition. The IDB, which uses a modular system of computer codes, also provides projections based on anticipated growth rates, schedules for new facilities, waste generation factors, and treatment assumptions. Projections for commercial waste are based on official DOE/Energy Information Administration (EIA) electrical growth projections.

The annual report of the IDB Program²⁹ presents summary-level tables and figures that compare the volumes and radioactivity levels of all forms of spent fuel and radioactive waste [high-level waste (HLW), transuranic (TRU) waste, and low-level waste (LLW)]. These materials are generated by commercial fuel cycles (including decontamination and decommissioning (D&D) activities), defense programs, remedial action programs (RAPs), and institutional and industrial operations.

Figure 2.2 illustrates the primary sources of data that are included in the IDB. A major program function is to translate various inputs from the numerous sources into a common and internally consistent data base. Data are collated and integrated to ensure that all forms are counted only once.

The IDB Program encompasses three major technical areas: data collection and processing, calculation of isotope generation and depletion, and data base development and user access. The steps involved in the flow of data from input to output, including data processing, are shown in Fig. 2.3.

29. *Integrated Data Base for 1986: Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-0006, Rev. 2, prepared by ORNL for U.S. DOE, September 1986.

ORNL DWG 87-581

Fig. 2.2. Sources of IDB data.

ORNL DWG 87-582

Fig. 2.3. Data manipulation in the IDB Program.

A formal system is in operation to assure the necessary quality control for both summary and detailed levels of data. These procedures were developed in cooperation with the DOE sponsors and the IDB Steering Committee. The quality control procedures are based on the requirements of the users of the data. A commitment has been made to document all actions. This documentation produces a record of events and assures that library files, published data, and computerized files are internally consistent and accurate.

Summary Data Tables and Figures

The usefulness of the IDB is inherent in its summation tables and figures, in which data for all waste forms are listed and displayed in a self-consistent manner. As an example, Table 2.2 shows the summary of spent fuel and waste inventories through December 31, 1985; and Table 2.3 includes projections to 2000, 2010, and 2020. These tables were generated from the core data files through the use of Statistical Analysis System (SAS) programming.

Table 2.2. Spent fuel and radioactive waste inventories as of December 31, 1985*

Spent fuel	Mass (MTU)	Activity (MCi)	Thermal power (kW)
BWRs (28,033 assemblies)	5,095	4,329	15,600
PWRs (18,319 assemblies)	7,762	9,940	37,300
High-level waste	Volume (m ³)	Activity (MCi)	Thermal power (kW)
SRP (DOE)	122,700	841	2,538
ICPP (DOE)	10,100	69	210
HANF (DOE)	222,000	517	1,573
WVDP (commercial)	2,310	32	96
Transuranic waste	Volume (m ³)	Activity (MCi)	TRU elements (kg)
DOE, buried	194,000	0.34	1,480
DOE, stored	92,100	1.58	1,755
Low-level waste	Volume (m ³)	Activity (MCi)	Land used (ha)
DOE sites	2,180,600	11.91	128
WVDP (closed 1975)	75,310	0.47	4
MFKY (closed 1977)	135,280	0.93	10
SHEF (closed 1978)	88,334	0.02	4
BARN	501,717	1.87	22
BETY	98,639	0.22	12
RICH	260,668	0.99	21
Commercial uranium mill tailings	Volume (m ³)	Activity (MCi)	Thermal power (kW)
At mill sites	99,958,000	0.87	19
Remedial action programs (DOE)	Volume (m ³)		No. of sites
UMTRAP (Uranium Mill Tailings)			25
FUSRAP (Formerly Utilized Sites)	118,920		29
SFMP (Surplus Facilities Management Program)	24,910		320
GJRAP (Grand Junction)	52,070		598

*Abbreviations used in this table are as follows: BWRs—boiling-water reactors; PWRs—pressurized-water reactors; SRP—Savannah River Plant, Aiken, S. C.; ICPP—Idaho Chemical Processing Plant, Idaho Falls, Idaho; HANF—Hanford Reservation, Richland, Wash.; WVDP—West Valley Demonstration Project, West Valley, N. Y.; MFKY—Maxey Flats, Ky.; SHEF—Sheffield, Ill.; BARN—Barnwell, S. C.; BETY—Beatty, Nev.; RICH—Richland, Wash.; RAP—Remedial Action Program.

Table 2.3. Current and projected volumes of spent fuel and radioactive wastes^a

Source of material and type	End of CY 1985		End of CY 2000		End of CY 2010		End of CY 2020	
	Volume (10 ³ m ³)	Radioactivity (10 ⁶ Ci)	Volume (10 ³ m ³)	Radioactivity (10 ⁶ Ci)	Volume (10 ³ m ³)	Radioactivity (10 ⁶ Ci)	Volume (10 ³ m ³)	Radioactivity (10 ⁶ Ci)
<i>DOE/Defense</i>								
HLW								
Interim storage	355	1,427	343	1,605	366	1,404	374	1,434
Glass	0	0	2.8	284	3.9	429	4.6	486
TRU waste								
Buried	194	0.3	194	0.3	194	0.3	194	0.3
Stored	92	1.8	182	3.5	243	4.7	303	5.9
LLW	2,181	11.9	4,043	18.3	5,159	20.9	6,256	22.9
UMTRAP and GJRAP								
Mill tailings	52		15,110		15,110		15,110	
Other waste	0		8,241		8,241		8,241	
FUSRAP	119		870		870	870		
SFMP								
TRU waste	1.3		3.3		3.4		3.4	
LLW	24		791		800		800	
Mill tailings	0		1,617		1,617		1,617	
<i>Commercial</i>								
Spent fuel (10 ³ MTIHM)	12.9	14,269	42.5	31,624	67.9	47,612	107.8	72,020
Commercial HLW glass (WVDP)			0.2	22.5	0.2	17.8	0.2	14.1
LLW	1,160	4.6	2,441	8.5	3,545	11.3	4,972	14.8
D&D (LLW)			0.6		91.3		795.9	
Mill tailings	99,958	0.9	146,480	1.3	197,303	1.7	265,865	2.3

^aAbbreviations used in this table are as follows: HLW—high-level waste; TRU—transuranic; LLW—low-level waste; UMTRAP—Uranium Mill Tailings Remedial Action Program; GJRAP—Grand Junction Remedial Action Program; FUSRAP—Formerly Utilized Sites Remedial Action Program; SFMP—Surplus Facilities Management Program; MTIHM—metric tons of initial heavy metal; WVDP—West Valley Demonstration Project, West Valley, N. Y.; D&D—decontamination and decommissioning.

Figure 2.4 presents the volumes of commercial and DOE/defense wastes and spent fuel accumulated through 1985 in the form of a pie chart that provides a clear pictorial overview of the relative amounts of this country's radioactive wastes. It is apparent that, on a volume basis, LLW dominates the total, with DOE/defense LLW comprising more than half this country's total waste. In Fig. 2.5 the radioactivities of the same wastes are presented in a similar fashion. On this basis, spent fuel comprises more than 90% of the radioactivity to be disposed of. It is obvious from these two figures why spent fuel and LLW are the major areas for concern in recent discussions on the storage and disposal of radioactive waste.

Waste-Specific Data

The capability of the IDB to produce numerous figures, maps, and charts directly from information within its core data files has recently been upgraded. The following eight figures represent a spectrum of the type of data that can be directly produced within the IDB Program. Figure 2.6 presents the projected mass of annual commercial spent fuel discharges. From this projection, it can be seen that, even though the number of new reactors coming on-line has been steadily decreasing, the trend for annual spent fuel discharges is toward an increase through the year 2020. Figure 2.7 also addresses spent fuel discharges, plotting the cumulative mass of the discharged fuel vs time and introducing the fuel age as a parameter. In 1985, very little fuel was more than 10 years old.

By the year 2000, about 40% will be more than 10 years of age; and by 2020, this fraction will have increased to ~60%.

The distribution of HLW volume and radioactivity by site is illustrated in Fig. 2.8. This bar chart indicates that, although the Hanford Reservation has the majority of the HLW volume, the largest amount of radioactivity is located at the Savannah River Plant. This discrepancy is due partially to Hanford's being an older facility, which would allow for additional decay time for some of its HLW, and partially to Hanford's having used, in its early history, a bismuth phosphate reprocessing scheme, which produced large quantities of dilute HLW. It is interesting that the Savannah River Plant will be the first DOE/defense site to vitrify its HLW into glass logs suitable for disposal in the spent fuel and HLW geologic repository.

Figure 2.9 gives a breakdown of the total volume of the retrievably stored TRU waste by site. This material, which includes both the contact- and remote-handled TRU waste categories, will be sent to the Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico. Idaho National Engineering Laboratory (INEL) is the major storage site for TRU material because it receives the bulk of the Rocky Flats weapons-related TRU waste.

A breakdown of the volume of DOE/defense LLW that is buried at the various DOE sites is shown in Fig. 2.10. This material is normally produced and buried on-site without shipment to a remote site. The DOE sites exhibiting the largest volumes of buried DOE/defense LLW are Hanford, Savannah River, and Oak Ridge. A similar

Fig. 2.4. Volumes of commercial and DOE/defense wastes and spent fuel accumulated through 1985.

ORNL DWG 86-14767

Fig. 2.5. Radioactivities of commercial and DOE/defense wastes and spent fuel accumulated through 1985.

Fig. 2.6. Projected mass (in metric tons of initial heavy metal) of annual commercial spent fuel discharges for the DOE/EIA Upper Reference Case.

breakdown for the buried commercial LLW is depicted in Fig. 2.11. West Valley, Maxey Flats, and Sheffield are all closed to additional waste, which leaves only Beatty, Richland, and Barnwell to receive commercial LLW until the various regional compacts (voluntary groupings of states) can implement the selection of their individual disposal

sites. Figure 2.12 illustrates the volume of LLW shipped to the commercial burial sites, by state, in 1985. Figure 2.13 projects the cumulative volume of both DOE/defense and commercial LLW generated until the year 2020. The DOE/defense share of the total LLW inventory is expected to drop slowly from ~65% in 1985 to ~55% in 2020.

ORNL DWG 86-14766

Fig. 2.7. Projected cumulative mass (in metric tons of initial heavy metal) of commercial spent fuel discharges for the DOE/EIA Upper Reference Case.

Special Studies and Requests

Special requests for information and special studies can be divided into three general categories:

1. ad hoc inquiries that can be responded to directly from existing IDB publications or prior data searches,
2. requests that require a special search of existing data bases or a new integration of existing data, and
3. queries that involve requests for special runs to generate new information.

Requests that can be responded to directly include historical annual storage rates, site-by-site volumes, and reconciliation of any apparent data omissions or duplications. Responses resulting from requested searches might include isotopic details of spent fuel discharges or the amount of heavy metal present in the TRU waste. Questions in these first two categories can usually be handled over the telephone. Queries in the third category which require special runs, are normally handled through DOE requests. Depending on the nature of the request, queries in this category might require a reimbursement of costs.

In summary, the IDB provides, in a wide variety of formats, technical information on spent fuel and radioactive waste in terms of inventories, projections, and characteristics. This information is useful in various program planning exercises and in

KEY TO ACRONYMS IN FIGS. 2.8 TO 2.11

BARN—Barnwell, South Carolina (commercial site)
 BETY—Beatty, Nevada (commercial site)
 BNL—Brookhaven National Laboratory, Brookhaven, New York
 HANF—Hanford Reservation, Richland, Washington
 ICPP—Idaho Chemical Processing Plant, Idaho Falls, Idaho
 INEL—Idaho National Engineering Laboratory, Idaho Falls, Idaho
 LANL—Los Alamos National Laboratory, Los Alamos, New Mexico
 LLNL—Lawrence Livermore National Laboratory, Livermore, California
 MFKY—Massey Flats, Kentucky (commercial site)
 NLO—National Lead of Ohio, Fernald, Ohio
 NTS—Nevada Test Site, Mercury, Nevada
 ORGDP—Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tennessee
 ORNL—Oak Ridge National Laboratory, Oak Ridge, Tennessee
 PAD—Paducah Gaseous Diffusion Plant, Paducah, Kentucky
 PANT—Pantex Plant, Amarillo, Texas
 PORT—Portsmouth Gaseous Diffusion Plant, Portsmouth, Ohio
 RICH—Richland, Washington (commercial site)
 SHEF—Sheffield, Illinois (commercial site)
 SNL—Sandia National Laboratories, Albuquerque, New Mexico
 SRP—Savannah River Plant, Aiken, South Carolina
 WVDP—West Valley Demonstration Project, West Valley, New York

Fig. 2.8. Distribution of volume and radioactivity of HLW by site and type through 1985.

ORNL DWG-86-14760

Fig. 2.9. Total volume of retrievably stored DOE/defense TRU waste through 1985.

Fig. 2.10. Total volume of buried DOE/defense LLW through 1985.

Fig. 2.11. Total volume of buried commercial LLW through 1985.

response to requests for data by DOE, state and local governments, and interested third parties. Ongoing efforts are concentrated on (1) making the data presented of consistently high quality, (2) streamlining the methodology for collecting data for the various waste types, and (3) using desktop publishing methods for simplifying the production of the annual inventory report.

2.7 WASTE IMMOBILIZATION TECHNOLOGY

The objective of the Waste Immobilization Technologies Program (WITP) is to provide technical assistance to DOE sites in developing and

applying appropriate, cost-effective processes to immobilize radioactive and hazardous chemical wastes. These processes are expected to result in reliable, feasible disposal schemes that comply with evolving regulatory requirements.

During this report period, the major programs included the following: (1) the development of pumpable grouts for Rockwell Hanford Operations' (RHO) Transportable Grout Facility (TGF); (2) the formulation of grouts for in situ stabilization of trenches at Idaho National Engineering Laboratory (INEL), ORNL, and Y-12; and (3) the support of operations for the hydrofracture facilities and Melton Valley storage tanks at Oak Ridge. These programs have met major milestones

ORNL-DWG 06-708

Fig. 2.12. Volume of LLW shipped to commercial burial sites, by state, in 1985.

Fig. 2.13. Historical and projected cumulative volume of LLW.

in resolving DOE's waste management challenges and have resulted in several publications.

Technical Support to RHO

The WITP continued to support Rockwell's TGF program for processing and disposal of Hanford

low-level wastes by (1) developing a reference formula and determining parameters and permissible deviations from the formula for neutralized cladding removal waste (NCRW), (2) developing a reference formula and defining operating parameters for phosphate-sulfate waste (PSW), and (3) performing preliminary grout formulation

studies for disposal of the double-shell slurry (DSS) waste stream.

With ORNL technical support, RHO is building the TGF for waste disposal in shallow-land vaults (Fig. 2.14). The TGF is scheduled for completion in December 1987. The use of cement-based grouting techniques is expected to provide a cost-effective, environmentally sound method for disposing of sludges, which make up a significant portion of the $1.78 \times 10^5 \text{ m}^3$ ($4.7 \times 10^7 \text{ gal}$) of waste at the Hanford site. The waste streams presently identified for processing in the TGF include NCRW, variable PSW, and high-nitrate, DSS waste. Accomplishments during this report period included the following:

1. a reference grout formula was finalized, and the ranges of blend component compositions and mix ratios using NCRW were optimized;
2. PSW reference blends were modified, and, based on relative amounts of phosphate/sulfate, preliminary formulations were selected; and
3. studies to select the most appropriate combination of dry-solids components for processing and immobilizing DSS were begun.

Cement-Based Grout Immobilization Studies on NCRW. Cladding removal waste (CRW) is a waste stream generated during the removal of the

protective cladding from spent fuel rods.^{30,31} Constituents of concern are cesium, zirconium, fluoride, and ammonia (typical initial concentrations of F^- and NH_4^+ in CRW are 3.11 M and 1.63 M, respectively). A reference formula was developed on the basis of experiments with neutralized CRW (NCRW) sludge in which a direct-strike method of neutralization was used. A major problem in grout formulation for this waste is that the fluoride acts as a set inhibitor. This effect was overcome by the addition of hydrated lime, $\text{Ca}(\text{OH})_2$, to inactivate the fluoride ion by forming calcium fluoride.

Grouts for NCRW disposal were tested with up to a 25% deviation from the reference formula and with variations in dry-solids/waste-mix ratios and in waste sludge dilutions. Varying the test conditions and parameters was necessary for gaging the effects of, and developing parameters for, actual operating variations. Testing confirmed that a grout having dry-solids components within a $\pm 5\%$

30. T. L. Sams, O. K. Tallent, T. M. Gilliam, and E. W. McDaniel, *Formulation Studies and Grout Development for Fixation of Cladding Removal Waste*, Milestone No. 181, ORNL/TM-9810, to be published.

31. T. L. Sams, E. W. McDaniel, R. D. Spence, and T. M. Gilliam, *Formulation Studies and Grout Development for Fixation of Neutralized Cladding Removal Waste*, Milestone No. 181—Part II, ORNL-6283, to be published.

ORNL DWG 84 1007A

Fig. 2.14. Schematic of Transportable Grout Facility planned at Hanford, Washington.

deviation from the reference formula (Table 2.4) will still meet all the specified performance criteria (Table 2.5).³²

Table 2.4. Recommended grout formula for NCRW disposal

Component	Amount	Range
Portland cement, Type III, wt %	44.8	42.8–46.8
Fly ash, Class F, Centralia, Washington, wt %	35.2	33.2–37.2
Indian Red pottery clay, wt %	7.6	6.0–9.2
Hydrated lime [$\text{Ca}(\text{OH})_2$], wt %	12.4	10.8–14.0
Mix ratio, kg/L (lb/gal)	0.78 (6.5)	0.72–0.84 (6.0–7.0)
Dilution of NCRW, vol %	60	50–70

Some properties of grouts prepared with the reference formula for disposal of NCRW are given in Table 2.6.

ANS 16.1 Leachability Indices from five replicate samples were 11.3 and 10.5 for ⁸⁵Sr and ¹³⁷Cs, respectively.

Cement-Based Grout Immobilization Studies on PSW. Phosphate-sulfate waste is composed of several RHO-generated streams and is essentially a variable mixture of phosphate and sulfate wastes from decontamination operations and from recharging ion-exchange columns, respectively.^{33,34} The concentrations of major constituents are given in Table 2.7.

Recent formulation development work has resulted in a blend that will tolerate variations in

Table 2.6. Properties of grouts using the reference formula for NCRW

Parameter	Measured value
Frictional pressure drop, kPa/m (psi/ft)	1.24 (5.5/100)
10-min gel strength, Pa (lb _f /ft ²)	14.4 (30/100)
28-d phase separation, vol %	0.0
Critical flow rate, ^a L/s (gal/min)	3.5 (56)

^aAt reference conditions.

Table 2.7. Concentrations of major components of PSW

Component	Concentration (M)
Na_3PO_4	0.15–0.5
Na_2HPO_4	0.01–0.02
Na_2SO_4	0.01–0.02
NaOH	0.01–0.02
NaNO_3	0.01–0.02

waste stream concentrations, particularly on the sulfate-rich side. When the new reference formula is coupled with the original formula developed for the phosphate-rich waste stream, all credible waste concentration scenarios can be made to meet all

32. T. M. Gilliam, E. W. McDaniel, L. R. Dole, and G. A. West, *Preliminary Data on Rheological Limits for Grouts in the Transportable Grout Facility*, ORNL/TM-9117, April 1987.

33. O. K. Tallent, E. W. McDaniel, and K. E. Dodson, *The Development of Multicomponent Hanford Facility Waste (HFW) Grout*, Milestone No. 173, ORNL/TM-10337, to be published.

34. T. L. Sams, E. W. McDaniel, C. L. Francis, and T. H. Keyes, *Preliminary Formulation Studies for Fixation of Variable Phosphate/Sulfate Hanford Facilities Waste*, Milestone 195, ORNL/TM-10291, to be published.

Table 2.5. Grout performance criteria for the TGF

Characteristic	Limiting value
Frictional pressure drop, kPa/m (psi/ft)	<2.5 (11/100)
10-min gel strength, Pa (lb _f /ft ²)	≤47.9 (100/100)
28-d phase separation, vol %	≤3
ANS 16.1 Leachability Index for ⁸⁵ Sr and ¹³⁷ Cs	≥6
Dry-solids/waste-mix ratio, kg/L (lb/gal)	<1.08 (9)
Critical flow rate, L/s (gal/min)	≤4.1 (65)
Major dry-solids blend components	Only components used in ORNL process
Dry-solids blend composition	Tolerates ±5% variation

performance criteria (Table 2.5). A primary concern with PSW has been the difficulty of simultaneously achieving minimal phase separation and critical flow rate requirements. This obstacle was overcome by using an admixture and by adjusting the blends.

Cement-Based Grout Immobilization Studies on DSS Waste. Double-shell slurry at RHO results from concentrating a number of dilute, noninteractive wastes.³⁵ It is characterized by high concentrations of sodium hydroxide (up to 13 M) and aluminum nitrate, with lesser concentrations of up to 24 other inorganic and organic constituents.

Exploratory studies were begun in order to develop a grout formulation for DSS immobilization that meets specifications as defined by process constraints with the TGF. Several aspects of this waste stream make the development work unique: (1) DSS grouts are mixed and cured at an elevated temperature, currently 60°C, to simulate actual conditions for this waste; (2) technetium tracer is used in selected tests to build a data base for evaluation of technetium leachability; and (3) in the dry-solids blends, ground-blast furnace slag is used instead of the usual fly ash and/or cement to provide a reducing environment for stabilizing the highly mobile technetium ion. The high ionic strength of DSS, due to high sodium and nitrate concentrations, tends to increase technetium leaching. It is expected that the use of ground-blast furnace slag will alleviate the leaching problem by preventing formation of the highly soluble pertechnetate ion; however, the use of slag tends to produce an undesirably high gel strength. Future work will be centered on optimizing the ratios of dry-solids blend components and then tailoring the grout, using an admixture, so that all performance criteria (Table 2.5) will be met.

Data Management and Quality Assurance

The experimental approach to grout formulation and the analysis of experimental results were modeled after guidelines provided by the American Society for Quality Control. An initial matrix of experimental runs was set up so that all reasonable combinations of variables and their responses could be studied. A large data base was required for accuracy of results; the experimental design included generation of numerical data in triplicate

for each combination of variables in the matrix. The data were recorded in technical notebooks and on computer files for subsequent analysis.

In addition to standard methods of data analysis, such as the *t*-test or hypothesis testing, response surface methodology and extreme vertices design were used to optimize the process with respect to the important variables. After an initial statistical workup, the data population was narrowed and an operating window for each variable was determined. Laboratory and statistical tests were then conducted on every reasonable combination of variables within the operating window until the optimum formulation was established.³¹

Product quality is further assured by control of materials, instrument calibration, training and certification of personnel, internal and external audits, peer review, documentation, and reporting.

Technical Support to In Situ Stabilization of Shallow-Land Burial Trenches

INEL. The WITP group has been developing grout formulations for in situ stabilization and long-term management of low-level and transuranic (TRU) wastes presently in shallow-land burial trenches at INEL.³⁶ In addition, the WITP group has prepared a cost estimate for grouting two experimental pits simulating INEL's TRU trenches and using ORNL-recommended grouts.³⁷ The acceptabilities of soil grouts, ordinary-particulate grouts, and fine-particulate grouts were evaluated on the basis of phase separation, compressive strength, freeze/thaw, penetration resistance, rheological, water permeability, column, and other tests. Soil grouts with soil-to-cement weight ratios from 0.91 to 1.60 were found to be suitable for open trench or drum disposal. Ordinary-particulate grouts containing Type I,II Portland cement; Class C fly ash; bentonite; water; and a fluidizer were formulated to fill large voids

35. T. L. Sams, O. K. Tallett, and E. W. McDaniel, *Preliminary Results of Grout Formulation Studies with Double Shell Slurry*, Milestone 59, ORNL/TM-9824, to be published.

36. O. K. Tallett, T. L. Sams, T. Tamura, T. T. Godsey, C. L. Francis, and E. W. McDaniel, *Grout Testing and Characterization for Shallow-Land Burial Trenches at the Idaho National Engineering Laboratory*, ORNL/TM-9881, October 1986.

37. R. D. Spence, *Cost Estimate of Grouting the Proposed Test Pits at INEL Using the ORNL Recommended Grouts*, ORNL/TM-10025, to be published.

within the soil/waste matrix of a closed shallow-land burial trench. Fine-particulate grouts containing fine cement (mean particle size, 9.6 μm) and water were formulated to fill smaller voids and to establish a grout-soil barrier to prevent water intrusion into the grouted waste trench. Solution, or chemical, grouts were evaluated as possible substitutes for the fine-particulate grouts, but their suitability in INEL soil has not yet been determined.

Performance requirements for lance injection or jet grouting of the trenches are listed in Table 2.8.

All requirements were met by three fine- and three ordinary-particulate mixes, and all criteria but the 10-min gel strength (not attempted because of this grout's thickness) were met by seven soil grout mixes.

For purposes of grouting the test pits at INEL, the two recommended grout compositions shown in Table 2.9 were chosen because they are the most economical of those tested satisfactorily. The soil grouts were judged insufficiently fluid for use in grouting the test pits. For a total void volume of 72.8 m^3 (2,570 ft^3), ~38.0 m^3 (10,040 gal) of coarse grout and 26.3 m^3 (6,950 gal) of fine, colloidal grout will be required. The total cost to INEL for double-grouting the two design pits is estimated to be \$100,000 using lance injection grouting and \$85,000 using jet grouting. Each technique has advantages, and it was suggested that both be tried as part of the test.

ORNL, SWSA-6. The WITP assisted the Environmental Sciences Division (ESD) in evaluating the effectiveness of particulate grouts as a remedial action alternative for buried low-level waste (LLW) by formulating a suitable grout and injecting it into Trench 150 in Solid Waste Storage Area-6 (SWSA-6).³⁸⁻⁴⁰ The purpose was to demonstrate that field-scale grouting with a particulate grout could be done economically and could achieve the desired objective of preventing sub-

Table 2.9. Recommended grout composition for stabilization of INEL trenches

Component	Amount
<i>Coarse grout</i>	
Water and fluidizer, wt %	35
Type I,II Portland cement, wt %	35
Class C fly ash, wt %	25
Beatonite, wt %	5
Fluidizer, wt %	0.5
<i>Fine grout</i>	
Colloidal cement, kg/L (lb/gal)	1.0 (8)
CFR-1 fluidizer, wt %	0.02

sidence and radionuclide migration. Shallow-land burial (SLB) of LLW commonly has these two problems. Past demonstrations have illustrated the value of solution grouting as a remedial action; this demonstration illustrated the value of particulate grouting.

Accordingly, ESD selected the candidate SLB trench (Trench 150) for the demonstration and characterized this trench and its contents prior to the grout injection. This characterization included reviewing the records of the waste placed in the trench prior to its being closed and backfilled in 1975. In addition, the groundwater and seepages were monitored and sampled for several years for this burial ground, particularly for the Trench 150 area several months prior to grout injection. A

38. R. D. Spence, T. Tamura, *Task Plan for Grout Injection/Evaluation Trench 150 SWSA-6*, ORNL/RAP-86/46, June 30, 1986.

39. R. D. Spence, *Grout Injection of Trench 150—ORNL SWSA-6*, ORNL/RAP-86/79, Letter Report, Sept. 30, 1986.

40. O. K. Tallent, E. W. McDaniel, R. D. Spence, and T. T. Godey, *Initial Formulation Results for In Situ Grouting of a Waste Trench at ORNL Site No. 6*, ORNL/TM-10299, January 1987.

Table 2.8. Performance criteria for in situ grouting at INEL

Parameter	Limiting value
7-d drainable water, vol %	0
28-d compressive strength, MPa (psi)	≥ 0.35 (50)
Compressive strength after freeze/thaw, MPa (psi)	> 1.4 (200)
Hydraulic conductivity, cm/s	$\leq 1 \times 10^{-7}$
10-min gel strength, Pa (lb/ft ²)	≤ 48 (100/100)
Shrinkage during curing, vol %	≤ 1

companion trench (Trench 152) was also characterized to act as a control comparison after Trench 150 was grouted.

An important part of this characterization effort was a water injection test of Trench 150 to estimate its accessible void volume and to establish its hydraulic behavior prior to grouting. The accessible void volume was estimated at 20%.

A grout formula was developed in the laboratory to meet the performance criteria of Table 2.10.

Table 2.10. Performance criteria for in situ grouting of ORNL Trench 150

Parameter	Limiting value
Apparent viscosity, mPa·s (cP)	<50 (50)
10-min gel strength, Pa (lb/ft ²)	<48 (100/100)
28-d phase separation, vol %	0
28-d compressive strength, MPa (psi)	≥0.41 (60)

Acceptable grouts were prepared at 12, 13, and 15 lb of dry blend per gallon of water for two dry blend compositions (see Table 2.11). These ranges in composition and mix ratio gave a wide operating window for the demonstration. In the grout injected, an average of these two blends was used to allow for blending variability, with a mix ratio target of 12.5 lb/gal for maximum fluidity. The two laboratory blend compositions and the field blend composition are given in Table 2.11. A fluidizer and set retarder (CFR-1), equivalent to 0.02 wt % of the dry blend, was added to the water.

Table 2.11. Dry blend compositions of grout formulations for injection of ORNL Trench 150

Component	Blend 1	Blend 2	Field blend
Type I Portland cement, %	36	41	39
Eastern Class C fly ash, %	58	54	55.5
Bentonite, %	6	5	5.5

The grout injection was done by adopting a simple injection technique, using ORNL employees as the workers, renting the necessary equipment from a local grouting firm, and using the engineering guidance of the Chem Tech grout development staff. The grout injection was done on time and under budget, with cooperation among several divisions, including Plant and Equipment, Operations,

Environment and Safety, Engineering, Environmental Sciences, and Chem Tech.

The dry blend components were stored and blended at the New Hydrofracture Facility at ORNL. Mixing the grout was accomplished by measuring a set quantity of water into a cement-mixing truck and blowing a preweighed quantity of blend into the truck drum as it rotated. The grout was kept slurried by the rotating drum until it was needed. A Moyno pump injected the grout into the trench from two 70-gal stirred tanks via a 2-in.-diam Schedule 40 pipe. Grout was added to the stirred tanks from the cement truck as needed. The 2-in.-diam pipes were inserted into the trench at predetermined points using a portable air-driven hammer. Grout was injected at a given location until refusal (pump rate too low or injection pressure too high) or until grout broke through to the surface. Pumping was continued in the same hole at a different depth unless breakthrough had occurred. About 30.3 m³ (8,000 gal) of grout was injected into Trench 150 through 36 holes in about 7 workdays. About 80% of this grout was injected into only 5 holes.

Trench 150 was 17 m long, 2.8 m wide, and 3.3 m deep after removal of some overburden to provide a collection basin for grout oozing and breakthroughs. Thus, the 30.3 m³ of injected grout filled about 19% of the total 157-m³ trench volume (about 97% of the estimated accessible void volume).

In conclusion, the demonstration was a success from an implementation standpoint. An inexperienced crew grouted practically all the accessible voids of an LLW trench in a reasonable amount of time at a reasonable cost. The ultimate effect on the trench's performance is currently being evaluated in the continuing monitoring and sampling program of the ESD. Some effects such as trench hydraulic behavior and grout placement will be known relatively quickly. However, ⁹⁰Sr migration is quite slow, and the effect of grouting on it will not be known for several years.

Y-12 Plant, UNC Disposal Site. The United Nuclear Corporation (UNC) Cold Scrap Recovery Facility, located at Wood River Junction, Rhode Island, was involved in the recovery of enriched uranium from scrap materials generated primarily by the defense program activities of DOE and its predecessor agencies. In August 1980, the UNC facility operations were shut down and UNC was required to decontaminate its facilities and lagoon

systems and remove the resulting low-level radioactive waste from the state of Rhode Island. DOE accepted the waste for disposal at its Y-12 Plant since the waste was a result of its defense program activities and there was a lack of capacity at commercial waste disposal facilities.

The UNC disposal site is located south of the Y-12 Plant on the crest of Chestnut Ridge. The site is an unlined pit that was cut out of the existing hillside and excavated down to planned grades (maximum cut of 9.75 m), with ~24 m of soil between the site and the top of the bedrock layer. The depth to the groundwater level is ~30.5 m. The area of the pit is ~3,700 m², and its depth ranges from 9 m on the north side to 0 m on the south side. This site was not equipped with leachate or runoff collection facilities.

Sixty crates, 238 boxes, and 29,002 drums containing waste have been stacked in the pit. This waste was generated during the decommissioning of the UNC facility in Rhode Island and includes sludge, soil, construction rubble, parking lot pavement, and office equipment, as well as liquid waste that was stabilized in concrete. The waste contains high levels of nitrates. Each container holds <4 g of uranium, yielding a total of 17,808 g of uranium at the site, of which 15,063 g is ²³⁵U.

During this report period, the WITP provided project management support for developing closure options for the Y-12 UNC disposal site. This included the coordination of efforts of both private contractors and Martin Marietta Energy Systems' personnel.

Samples taken from randomly chosen drums containing contaminated soil and from drums containing solidified sludge were analyzed for Resource Conservation and Recovery Act (RCRA) hazardous waste characteristics by the Analytical Chemistry Department, Quality and Technical Services Division, Oak Ridge Gaseous Diffusion Plant. Results of the analysis show that the soil samples are noncorrosive while the solidified sludge is very close to the RCRA upper limit of 12.0 pH. For both the soil and sludge samples, the levels of the eight Extraction Procedure (EP) Toxicity metals (As, Ba, Cd, Cr, Hg, Pb, Se, and Ag), herbicides, and pesticides are well below the U.S. Environmental Protection Agency (EPA) maximum concentrations; and the concentrations of radionuclides present in the extract are also very low. On the basis of these analytical results, the waste can be classified as nonhazardous (as defined under RCRA), however, the concentration of nitrates present in the extract from the sludges

is very high (~50,000 ppm). In addition, the presence of ettringite, an expansive agent in the cement, has been confirmed and quantified by Michael Grutzeck, of the Materials Research Laboratory, Pennsylvania State University. The ettringite has already caused several of the drums at the UNC disposal site to swell and split open. The presence of ettringite and its ultimate control became a major issue in the development of a closure option.

Using site data and analyses of the UNC wastes and leachates available at the time, Roy F. Weston (a subcontractor) produced a preliminary site evaluation document. In this document, Weston proposed risk zones and did a preliminary pathways analysis using the Hydrologic Evaluation of Landfill Performance (HELP) Model developed by the U.S. Army Corps of Engineers Waterways Experiment Station in Vicksburg, Mississippi, and the WMPLUME Model, which is a subpart of the SOLUTE package developed by the Holcomb Research Institute in Indianapolis, Indiana. These pathways and preliminary risk assessments established the baseline performance of the no-action site option, against which the performances of the site closure options will be compared. The results of this analysis predicted that, under present site conditions, nitrate-contaminated leachate would reach the groundwater in ~10 years. In addition, the simulation predicted maximum nitrate concentrations (as nitrogen) of ~190 ppm in the groundwater and a range of approximately 10 to 30 ppm in the surface water, depending on the extent of groundwater discharge to the stream.

On the basis of the results of the preliminary pathways analysis, two site closure options have been identified: placing a simple clay cap over the site or placing a more complex RCRA cap over the site. In addition, to prevent horizontal infiltration of groundwater, construction of a barrier on the east side of the site has been proposed, in conjunction with the construction of one of the two caps. Pathways analyses are currently being prepared for the two site closure options, using the data and models used in the preliminary site evaluation, so that the closure options can be compared with the no-action option.

Technical Support to Y-12 and K-25: Grout Formulation for Immobilization of Mixed Organic-Containing Wastes

Work has been completed on the formulation of a grout to immobilize organic-containing waste

from the 1,893-m³ (500,000-gal) F-1 tank at Y-12.⁴¹ The project is demonstrably feasible. In mid-FY 1986, formulation studies were redirected toward the immobilization of drummed K-1232 waste (at K-25) that has a relatively massive organic content (total organic carbon and total phenol). Both the F-1 and the K-1232 waste grouts must meet the performance criteria of the K-25 batch facility, which are (1) no drainable water at the time of disposal, (2) acceptable leachability, and (3) a minimum 28-d unconfined compressive strength of 2.07 MPa (300 psi).

The F-1 tank sludge consists of mixed organic- and inorganic-containing biodegraded waste. The development work was primarily concerned with fixation of the organic components since previous formulation work had produced grouts that would readily immobilize the metallic components. Experimental results indicated that a dry-solid blend of Type I Portland cement, Class F fly ash, bentonite clay, and Micro-Cel E (a calcium silicate additive) would be effective in the retention of organic and metallics. It had been found that retention of total phenols within a cementitious matrix was sharply improved by silicate addition; however, blends containing sodium silicate encountered a surface-cracking problem upon exposure to ambient air. Micro-Cel E was almost as effective and showed no cracking problem.

Some uncertainty exists about the effectiveness of bentonite clay with this waste. Early tests indicated no deleterious effect from its exclusion, but later tests were less conclusive. In future work, Y-12 grouting and leaching studies will be performed with F-1 waste (20 to 25% solids) from a filtration process. After the optimum blend is defined, several demonstration pours to form large grout monoliths will be made at the K-25 batch plant.

Formulation of an acceptable grout using the K-1232 waste is proving much more difficult, probably because of its high phenolic content. Micro-Cel E has been extremely ineffective in preventing leaching of the phenols of this sludge. In addition, chemical analysis of the sludge has been problematic. The samples are difficult to characterize, partly because of the hydrolysis of certain phenolic compounds into various nitrated phenols. Additional development work needs to be performed before individual assays on this type of material can be considered reliable.

Hydrofracture Support

Pursuant to the development of a comprehensive closure plan for the hydrofracture facilities at ORNL, the WITP has developed a task plan⁴² for testing representative hydrofracture grouts for leachability and physical integrity as a measure of their potential for successful long-term sequestering of pollutant species.⁴³

The hydrofracture process was used for a number of years at ORNL. It was a method for disposing of hazardous and radioactive liquid wastes by mixing the waste with cement containing various additives and pumping the mixture under high pressure into shale beds at underground depths of ~300 m. The last injections were made in 1984. Since that time, the decision was made to discontinue use of the hydrofracture process. This decision dictated the need for development of a comprehensive closure plan for the facilities, which would include monitoring and experimental activities designed to allow prediction of potential long-term threats to the environment posed by the presence of waste-bearing hydrofracture grouts in the deep shale beds.

The WITP task plan⁴² for grout durability studies addresses both the chemical and physical integrity of representative hydrofracture grouts. Chemical integrity or leachability will be determined by a set of leach tests designed to predict the long-term resistance of grouts to leaching when exposed to a high-chloride (>130,000-ppm) brine, which is the typical liquid environment in the shale beds at hydrofracture depths. Leachates will be analyzed for a wide range of chemical constituents, including the EP Toxicity metals As, Ba, Cd, Cr, Hg, Pb, Se, and Ag; Cs, Sr, and Co; NO₃⁻; and the cementitious matrix elements Ca, Mg, K, Fe, Al, and Na. Such analyses will provide source term

41. J. E. Mrochek, T. M. Gilliam, and E. W. McDaniel, "Grout Formulation for the Immobilization of Mixed Organic-Containing Wastes," presented at the American Nuclear Society International Meeting on Low-Intermediate- and High-Level Waste Management and Decontamination and Decommissioning, Niagara Falls, N. Y., Sept. 14-18, 1986; proceedings to be published in 1987.

42. M. E. Reeves and T. M. Gilliam, *Proposed Grout Durability Studies in Support of the Remedial Investigation of the Hydrofracture Operations at Oak Ridge National Laboratory*, ORNL/RAP-15, April 1987.

43. T. M. Gilliam and J. A. Loflin, *Leachability Studies of Hydrofracture Grouts*, ORNL/TM-9879, November 1986.

data on the pollutant species, as well as an indication of the cement matrix integrity of the grouts after long periods of immersion in brine.

Physical tests proposed for hydrofracture grouts include compressive strength, density, porosity and internal surface area, permeability, and microscopic examination. These tests serve essentially two functions: (1) They provide descriptive, baseline information on physical characteristics of hydrofracture grouts. (2) Since all physical tests will be performed on both leached and unleached samples, they allow predictive models of grout durability to be constructed based on observations of the effects of exposure to deep brine on the physical structure and characteristics of representative hydrofracture grouts.

Some preliminary work assessing hydrofracture grout durability and chemical integrity has been completed. Experiments were performed which assessed the leachabilities of ^{90}Sr , ^{137}Cs , As, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, and Sb from hydrofracture grouts exposed to the following leachants: distilled water, a high-carbonate shallow groundwater, and a high-chloride deep brine. The shallow groundwater was representative of site groundwater above the shale formation, while the deep brine was representative of site shale formation waters. The ANS 16.1 Leachability Indices obtained in all three leachants are shown in Table 2.12.

As indicated by the data, deep brine is the most aggressive of the three leachants studied. Evidence has been presented which indicates that the chloride salts contained in the brine penetrate into

the grout to alter the pore-water chemistry and control the release of strontium and cesium. Two by-products of the chloride pore-water reactions, CaCl_2 and $\text{Mg}(\text{OH})_2$, do not appear to be migrating from the grout. Accumulation of these salts will plug the grout pore voids; therefore, the aggressive action of the chlorides may be self-limiting. Nonetheless, on the basis of these data, deep brine was chosen as the reference leachant for the continuing grout durability studies.

In addition to these studies, leach studies were performed which demonstrated that the grouts were not EP Toxic, as defined by the RCRA. Significantly, these efforts led to the development of a relationship between EP Toxicity results and the ANS 16.1 Leachability Indices for pumpable grouts. The general relationship is as follows:

$$\frac{C}{C_o} = \frac{(31)10^{-\bar{L}_i}}{M + \rho_w} \quad (1)$$

where

- C = EP Toxicity leachate concentration, mg/L;
- C_o = original waste concentration, mg/L;
- L = ANS 16.1 Leachability Index;
- M = grout mix ratio, kg/L;
- ρ_w = density of original waste, g/cm³.

Although the EP Toxicity procedure requires an acetic acid solution as the leachant, experience at ORNL has shown that, generally, results from

Table 2.12. Average ANS 16.1 Leachability Indices obtained with various leachants, using grouts cured for 24 d

Constituent	$\bar{L}_i$ in distilled water	$\bar{L}_i$ in high-carbonate groundwater	$\bar{L}_i$ in deep brine
^{137}Cs	10.6	10.5	8.8
^{90}Sr	10.6	11.7	8.6
As	13.6	12.7	11.3
Ba	13.1	12.0	8.9
Cd	17.3	17.1	10.9
Co	13.9	13.9	10.5
Cr	13.8	13.4	11.4
Cu	15.1	14.3	10.7
Hg	13.4	13.3	8.1
Ni	15.0	15.1	11.1
Pb	15.3	15.3	10.3
Sb	13.1	12.1	9.7

leaching in distilled water are similar for grouts that pass the structural integrity test. Therefore, the leachability index used in Eq. (1) can be obtained using distilled water, the ANS 16.1 standard leachant. This relationship can be used as a powerful tool for quality assurance during disposal operations to guide formulation development efforts and to expand existing data bases to a wider variety of mixed wastes.

ORNL Pond Sediment Stabilization Study

The closure of several ORNL impoundments has been investigated as part of the Environmental Restoration and Facilities Upgrade Program.⁴⁴ The eight impoundments considered in this study are located in two general areas: the 3500 area of the main ORNL complex and the 7900 area of Melton Valley. Three of the four impoundments located in the ORNL complex are currently active and receive process waste streams from the main part of the ORNL facility on Bethel Valley Road. These process waste streams are primarily effluents which contain traces of metals and solvents and little or no radioactivity under normal operating conditions but which could contain higher levels of radioactivity resulting from equipment failure or human error. The four impoundments located at the Melton Valley ORNL facilities serve as holding basins. Two serve as intermediate storage and collection basins for the High-Flux Isotope Reactor facility, while the other two receive process waste streams from the Transuranium Processing Plant and the Thorium-Uranium Recycle Facility. The sludges that have accumulated in the bottoms of these impoundments were generated primarily from the settlement of suspended solids, although precipitation of metals is a secondary source generator.

During this report period, WITP personnel performed an evaluation of the feasibility of stabilizing the sediments associated with the impoundments. The methodology used in this evaluation consisted of a literature survey, vendor contacts, determination of the regulatory criteria to be applied to the pond sediments, and the development of general criteria to be used in the comparison of waste technologies.

Using U.S. Nuclear Regulatory Commission (NRC) and EPA regulations and available data, we classified sediments contained in the impoundments. All the impoundments contain detectable

quantities of radioactive material. In the absence of accepted minimal levels, the sediments are assumed to be classified as radioactive wastes. According to NRC regulations, they are Class A wastes. However, a previous study indicated that transuranic concentrations in impoundment 3524 samples are within 6% of the threshold value for Class C wastes. In the absence of statistical information on the precision of these analyses, the 3524 sediments were assumed to be Class C wastes.

At the time of the study, there was insufficient documentation of the wastes contained in the impoundments to make a definitive judgment on the issue of "listed wastes" (40 CFR 261). Also, since the available analyses indicated no significant quantities of listed wastes, it was assumed that the sediments contained none. A parallel study conducted on seven of the impoundments indicated that they are not characteristically hazardous, while a previous study indicated that EP Toxicity leachate concentrations from impoundment 3513 samples exceed permissible mercury levels by an order of magnitude. As a result, it was assumed that impoundment 3513 is RCRA hazardous, while the other impoundments have been tentatively classified as RCRA 3004 U, requiring additional monitoring to determine whether there is any release of hazardous materials.

During the course of the literature survey and vendor contacts, four stabilization technologies (in asphalt, cement, glass, and polymers) and two basic implementation techniques (batch plant and in situ) were identified. The general criteria that were developed to compare the waste forms produced by the technologies under consideration were grouped under the following categories: vendor technology, waste form properties, and implementation techniques.

This study evaluated the use of sediment stabilization to support the site remedial action plan. The principal driving forces for stabilization are state and federal regulations governing waste classification and the resulting disposal requirements. The regulatory classifications of the sediments indicate that stabilization (1) is a viable method of rendering impoundment 3513 characteristically nonhazardous, (2) is required for the impoundment 3524

44. R. D. Spence, M. V. Helfrich, T. M. Gilliam, G. D. Del Cul, and T. T. Godsey, *Status Report on the Evaluation of Stabilizing Sediments of Selected ORNL Impoundments*, ORNL/TM-10473, to be published.

sediments, and (3) may not be required for the remaining six impoundments. The six ponds that have been classified as Class A waste need only meet the following waste form criteria: no free liquid and all liquids absorbed. Therefore, removal of free liquid from the sediments (by filtration, drying, or addition of water-sorbing materials) and disposal/storage of the remaining sediments on-site or elsewhere, such as in drums, was an option proposed by the study.

Using the general criteria developed for this study, we recommended this stabilization method: consolidation of the sediments followed by solidification using the cement batch-plant implementation technique. We also recommended that the impoundment water be drained and treated by the process water treatment system to reduce the overall cost and the volume of the final product. A letter report was drafted to document the evaluation procedure and recommend stabilization techniques. On the basis of the results of this study, a recommended site closure plan and a Request for Proposals from vendors were developed.

Melton Valley Storage Tank Support

At ORNL, ion-exchange processes used to remove primarily ^{137}Cs and ^{90}Sr , as well as trace amounts of ^{60}Co and rare earths, from building process wastes yield a mixed, low-level, nitric-acid-based waste. The acid is heated in evaporators so that as much as possible can be recovered for reuse in the ion-exchange circuit. Following acid recovery, the resulting slurry is made alkaline at approximately pH 12 using sodium hydroxide.

The alkaline sodium nitrate solution is then pumped to storage tanks on-site at ORNL, where it is stored in containers referred to as the Melton Valley storage tanks. The radioactive solution contains activity in the range of 98 to 980 MBq/L and ~30 wt % nitrate.

Concern over the rate at which eight 190,000-L storage tanks were filling prompted an investigation into those fixation media which were immediately available for on-site use and offered a good volumetric-reduction efficiency. Bitumen was then chosen as one of the possible fixation media that could potentially immobilize the first 378,000 L of waste.

The decision process used in choosing a fixation technology or vendor involves laboratory performance testing of prospective vendors' small-scale

waste form specimens. Performance testing includes compliance with test methods centered primarily around the NRC's 10 CFR 61 Branch Technical Position Paper, which recommends methods for demonstrating compliance with these waste form performance criteria. In addition, tests required by the EPA have been performed on the specimens.

During June 1986, waste form specimens were prepared at a vendor's site using a 53-mm, twin-screw extruder. These specimens were prepared in the presence of ORNL personnel to corroborate that conditions were as close as possible to those which might be encountered in the field, and then the specimens were transported to Oak Ridge for testing.

Because contamination of equipment and transportation of radioactive materials presented a major problem, a surrogate waste was prepared to represent the Melton Valley waste solution. The resulting chemical analysis of the as-prepared surrogate waste is presented in Table 2.13.

As shown in Table 2.13, the waste contained metals and nonmetals of concern to the EPA and the state of Tennessee, in addition to the major salt, sodium nitrate. Currently, the waste is expected to be classified as a mixed LLW, but "ultrafiltration" of the waste is expected to lower the concentration of many metals and nonmetals

Table 2.13. Surrogate waste composition representing the Melton Valley nitrate-based waste
(Density at 25°C = 1.25 g/cm³)

Species	Concentration (mg/L)
Arsenic	47
Barium	42
Cadmium	50
Calcium	13,000
Cesium	48.9
Chloride	2,200
Chromium	46
Lead	47
Magnesium	990
Mercury	38.4
Nitrate	238,000
Selenium	43
Silver	48
Sodium	77,000
Strontium	47
Sulfate	560

prior to waste fixation. Not all of these metals exist in the actual waste; however, we included them because of interest in the ability of bitumen to impede the leachability of all eight chemical species of current concern to the EPA (As, Ba, Cd, Cr, Hg, Pb, Se, and Ag). Although some of the chemical species occur in amounts ranging from only 5 to 30 ppm in the actual waste, analytical detection limit restraints dictated the use of at least 50 ppm for each of the EPA chemical elements.

During the demonstration in which the specimens were prepared, three target levels of waste loading were achieved: 40, 50, and 60 wt % salt.

Methodology. Waste Forms. Cylindrical waste form specimens were cast into the proper size and geometry to facilitate regulatory testing using aluminum molds with Neoprene bottoms. Aluminum sheets 10 mils thick were cut to form tubes, and aluminum-backed tape was used to hold the seams together. Disks molded from Neoprene were tightly fitted into the bottoms of the tubes, and a thin layer of silicone-based cement was placed on the edge of the disks to ensure a complete seal.

The aluminum molds were made longer than desired to compensate for the high thermal expansion coefficient of bitumen, which causes the surface to shrink inward upon cooling. However, typical fill efficiencies were >90% during full-scale operation. Following removal of the molded samples from the molds, the top of each sample was carefully cut and leveled using a razor knife.

During the demonstration, the extruded bitumen product exiting the extruder unit readily flowed into the aluminum molds. The forms were cooled and then placed in plastic bags for storage prior to laboratory testing.

Physical Testing and Characterization. Because 10 CFR 61 requires that waste forms exhibit minimum unconfined compressive strengths of 50 psi (0.34 MPa), compression testing was performed. The recognized unconfined compressive strength test for bituminous mixtures is the ASTM method D 1074-83. This test was performed on right-circular cylinders of bitumen waste forms molded in soft drink cans, with a height-to-diameter ratio of 1.

The vertical rate of deformation for these tests was set at 0.127 cm/min per centimeter of sample height. The NRC has recommended that, because bitumen flows rather than fractures, the unconfined compressive strength should be evaluated at

the point where 10% deformation in the specimen height occurs.

Leaching. Two kinds of regulatory leach tests were performed in this evaluation: the EPA's EP Toxicity test and the ANS 16.1 leach test recommended by the NRC.

The EP Toxicity tests were performed using acetic acid, according to the regulatory procedure, over a 24-h period. In this test, high-density polyethylene vessels were used; they were connected to an automatic acetic acid delivery system composed of a buret, a set-point controller, and an electric solenoid valve.

The leachates from the tests were analyzed for all waste species, not just the eight metals of current concern to the EPA. Analyses of the leachates from both leach tests were performed using only EPA-approved methods in an EPA-certified laboratory.

Additionally, the 90-d version of the ANS 16.1 leach test was performed on all waste forms. The leachant used in these tests was distilled water having an electrical conductivity of <5 $\mu\text{S}/\text{cm}$ (<5 $\mu\text{mho}/\text{cm}$) at 25°C and a total organic content of <3 mg/L (<3 ppm).

Leach indices resulting from the use of this leach test are reported for all species after 90 d even though this test was established primarily for radionuclides. In this way, the indices serve as a "figure-of-merit" that may be used for modeling the expected fractional release rate following disposal.

Results. Unconfined Compressive Strength. One of the first tests performed on the waste forms was the unconfined compressive strength test. The results of the tests are shown in Table 2.14, with the pure, salt-free bitumen sample serving as the blank.

The regulatory minimum compressive strength is 345 kPa (50 psi). As shown in Table 2.14, all waste forms easily passed this minimum, including

Table 2.14. Unconfined compressive strength data at 10% deformation

Salt loading (wt %)	Compressive strength (MPa)
0.0 (blank)	1.72
41.6	2.48
50.6	3.52
60.2	4.29

the blank. The greater resistance offered by increasing amounts of salt is evident from the upward trend in compressive strength. The waste form containing ~60 wt % salt was the maximum loading achievable during the demonstration.

Leaching—EP Toxicity Test. As the data in Table 2.15 show, all waste forms at all levels of loading easily passed the EP Toxicity test. The data show that, for most of the metals, the concentration in the leachates was at or near the analytical detection limit.

Leaching—ANS 16.1 Test. The NRC requires that a radionuclide possess a leach index of at least

6 when leached in accordance with the ANS 16.1 leach procedure. Because the leach index is the negative logarithm of the effective diffusion coefficient, it can be used in modeling the expected leaching behavior of any waste species if certain assumptions are made. The leach index then becomes a "figure-of-merit" used to ascertain the ability of a given waste form or treatment scheme to impede the leachability of a waste component.

The resulting leach indices obtained from the ANS 16.1 tests for all waste components are presented in Table 2.16. The data show that both cesium and strontium exceeded the NRC's

Table 2.15. Comparison of EP Toxicity test results as a function of salt loading

Species	Concentration (mg/L) at salt loading (wt %) of				
	26.1	41.6	50.6	60.2	Limit
Arsenic	0.008	<0.005	<0.005	<0.005	5
Barium	0.017	0.010	0.024	0.049	100
Cadmium	<0.0030	0.0032	0.0081	0.024	1
Chromium	<0.010	<0.010	<0.010	<0.010	5
Lead	0.010	0.033	0.010	0.016	5
Mercury	<0.0002	0.0003	0.0003	<0.0002	0.2
Selenium	<0.0050	0.027	<0.005	0.005	1
Silver	<0.0060	<0.0060	<0.0060	<0.0060	5
Nitrate	12.9	54	160	320	44.3

Table 2.16. Tabulation of average leach indices after 90 d of leaching in distilled water

Species	Leach index at salt loading (wt %) of					
	41.6		50.6		60.2	
	Average ^a	Standard deviation ^b	Average ^a	Standard deviation ^b	Average ^a	Standard deviation ^b
Arsenic	>10.9	0.8	>11.1	0.8	>11.3	0.8
Barium	10.7	0.5	10.2	0.4	9.8	0.3
Cadmium	>10.7	0.3	10.6	0.3	10.4	0.5
Calcium	10.5	0.4	9.8	0.3	9.4	0.3
Cesium	>8.0	0.4	>8.4	0.7	>8.6	0.8
Chloride	>9.7	0.7	>9.7	0.5	>9.6	0.4
Chromium	>10.3	0.8	>10.5	0.8	>10.7	0.8
Lead	9.7	1.1	10.0	0.9	10.2	0.9
Magnesium	10.7	0.5	10.5	0.5	10.4	0.5
Mercury	>13.7	0.8	>13.9	0.8	>14.1	0.8
Nitrate	10.6	0.5	9.8	0.2	9.3	0.3
Selenium	>10.9	0.8	>11.1	0.7	>11.2	0.7
Silver	>10.8	0.8	>11.0	0.8	>11.2	0.8
Sodium	10.4	0.4	9.5	0.2	9.2	0.3
Strontium	10.7	0.5	10.0	0.3	9.4	0.3

^aArithmetic average of three replicates.

^bRoot-mean-square of the standard deviations of the replicates.

minimum leach index of 6. When this limit is applied to all remaining species, even though they are not radionuclides, it is evident that they have also exceeded the minimum leach index by a large margin.

Conclusions. Regulatory performance testing of extruded bitumen has shown that the relatively viscous form of oxidized bitumen used in this testing program has been able to meet all currently required performance tests. Bitumen-based waste forms may be able to provide superior resistance to the leachability of the current EPA metals of concern while showing promising results for the problematic nitrate.

Polymer Impregnation of Cement-Based Waste Forms

A program to investigate the use of polymers in the sequestration of high-nitrate wastes stored in 190,000-L (50,000-gal) tanks on the ORNL site (referred to locally as the Melton Valley storage tanks) was begun during this report period. This waste stream is a low-level radioactive liquid waste composed primarily of 28 wt % sodium nitrate from neutralized nitric acid; the waste originates at ion-exchange processes on-site.

The concept of using polymers to aid in reducing the leachability of very mobile and problematic waste species such as tritium, technetium, or nitrate from cement waste forms is not new. The objective of polymer impregnation is to block the internal pores and channels of the cement form, thereby lowering the diffusivity of the waste components. Research into this area has been pursued by many investigators over the years, with limited success for several fundamental reasons.

Attempts at emulsifying the organic-based polymer with cement-based dry solids and waste have resulted in problems with set (i.e., slowing or halting of cement hydration reactions due to the presence of the polymer). Most attempts resulted in failure since the waste mixture never became a solid mass with the desired properties.

The utility of adding polymer to the surface of a cured waste form in hopes that some degree of penetration could be achieved was also found to be very limited. Since the pores contain some free water, the nonpolar polymer has difficulty diffusing to any useful depth, typically penetrating only a few millimeters in a 2- to 4-cm-diam laboratory-scale sample.

During this report period, initial work duplicated the findings of other investigators regarding the above limitations; however, recent work with a low-viscosity styrene monomer shows promise. When the monomer is combined with a modified, cement-based blend in conjunction with a benzoyl peroxide catalyst, the resulting polystyrene polymer has two significant properties: low viscosity and an ability, because of the polymer ring structure, to quench the free radicals produced by ionizing radiation. Several approaches to incorporating the polymer into a cement waste form are being investigated: (1) the addition of various emulsifying agents with rapid-set, water-clean, dry-solid blends, in an attempt to overcome the set retardation usually incurred when polymer is mixed directly into the cement matrix, and (2) the enhancement of polymer penetration into the waste form from the surface, aided by microwicking fibers. There have been some encouraging results. Work is continuing, and results will be presented during the next report period.

Conclusions

The development of pumpable grouts and grouts for in situ stabilization has been the major thrust of the WITP, with important development efforts being invested in projects for DOE's Hanford site, the INEL, and DOE's Oak Ridge installations. Quality assurance (QA) is an integral part of the program. QA procedures are reviewed and updated as the program requires and will continue to be emphasized in the development work.

The WITP will continue its commitment to transfer technologies to other DOE and federal agencies.

2.8 OTHER WASTE MANAGEMENT ACTIVITIES

Waste Classification

In 1984 the U.S. Nuclear Regulatory Commission (NRC) made available an informal draft, "Advanced Notice of Proposed Rulemaking (ANPR)," concerning the definition of high-level waste (HLW) pursuant to its responsibilities under the Nuclear Waste Policy Act (NWPA) of 1982. Promulgation of this rulemaking would have reclassified many existing transuranic wastes as

HLW, thus requiring much more extensive treatment and greatly diminishing the need for very expensive facilities already under construction. Additionally, the rulemaking did not appear to be based on scientific or radiation protection principles. As a consequence, the DOE Office of Defense Waste and Transportation Management requested Chem Tech take the lead in (1) providing technical assistance in commenting on the NRC draft and (2) developing an alternative definition of HLW that was based on scientific and radiation protection principles and had minimal adverse impacts on DOE operations.

During the past 3 years, staff members of Chem Tech and the ORNL Health and Safety Research divisions have worked at developing this alternative definition, which resulted in a comprehensive classification system for all radioactive wastes that met the criteria stated above and was acceptable to DOE operating contractors. This result, documented in a technical report,⁴⁵ also provided much of the basis for the NRC changing its ANPR position, as reflected in a recently published *Federal Register* notice.⁴⁶ A summary of the results of this activity is given in the following paragraphs.

The NWA provides the following definition of HLW:

(A) the highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and

(B) other highly radioactive material that the (U.S. Nuclear Regulatory) Commission, consistent with existing law, determines by rule requires permanent isolation.

As stated above, the primary purpose of Chem Tech's activities was to develop a generally applicable quantitative definition of HLW that addresses the description in paragraph (B) above while also encompassing paragraph (A). The development of a generally applicable definition of HLW also results in definitions of two other waste classes, which are called transuranic (TRU) waste and equivalent and LLW.

High-level radioactive waste traditionally has been defined in terms of its source as waste from chemical reprocessing of spent nuclear fuel, and a qualified source-based definition is given in paragraph (A) above. However, wastes from fuel reprocessing were recognized as having certain characteristics, related to short-term risks from waste

operations and to long-term risks from waste disposal, that provided the basis for the source-based definitions of HLW:

1. high concentrations of fission products, principally ⁹⁰Sr and ¹³⁷Cs, resulting in high heat generation rates and external radiation dose;
2. high concentrations of long-lived radionuclides, principally the alpha-emitting TRU radionuclides, that would result in high internal radiation dose per unit activity of inhaled or ingested material.

These characteristics have been used to develop a generally applicable definition of HLW and the other waste classes.

On the basis of the definition of HLW in paragraph (B) of the NWA and other historical precedents for defining such wastes, it is proposed that HLW is waste that (1) is highly radioactive and (2) requires permanent isolation. This conceptual approach results in a two-dimensional waste classification system in which one axis is related to the concept of "highly radioactive" and is associated with shorter-term risks and the other axis is related to the concept of "requires permanent isolation" and is associated with long-term risks from waste disposal.

The conceptual definition of HLW also leads to the following definitions of TRU waste and equivalent and LLW:

1. TRU waste and equivalent is waste that requires permanent isolation but is not highly radioactive;
2. LLW is waste that does not require permanent isolation, without regard to whether or not it is highly radioactive.

A generally applicable quantitative definition of "highly radioactive" has been developed by associating this concept with high levels of decay heat (power density) or external radiation dose in a manner consistent with the first characteristic of source-based HLW described above. On the basis of analyses of levels of power density and external radiation dose that limit system design or operation

45. D. C. Kocher and A. G. Croff, *A Proposed Classification System for High-Level and Other Radioactive Wastes*, ORNL/TM-10289 (in publication).

46. U.S. Nuclear Regulatory Commission, "Definition of High-Level Radioactive Waste," *Fed. Regist.* 52(39), 5992 6001 (Feb. 27, 1987).

in controlling short-term risks in a variety of activities, including waste disposal, the following generally applicable definition is proposed: "highly radioactive" means (1) a power density $>50 \text{ W/m}^3$ or (2) an external dose-equivalent rate, at a distance of 1 m from the waste, $>100 \text{ rem/h}$ (1 Sv/h). Thus, a determination of whether a waste is highly radioactive can be based on direct measurements of power density and external dose rate without knowledge of radionuclide concentrations in the waste. However, these levels of power density and external dose rate are also used to derive, on the basis of simple models and calculations, radionuclide concentrations that define a highly radioactive boundary. These boundary concentrations are given in Table 2.17 and can be used when the radionuclide concentrations in the waste are known. We note that the concentrations defining the highly radioactive boundary for ^{90}Sr and ^{137}Cs correspond to the limits for near-surface disposal of these radionuclides as Class C waste.⁴⁷

A generally applicable quantitative definition of "requires permanent isolation" is developed by associating this concept with high concentrations of long-lived radionuclides in a manner consistent with the second characteristic of source-based HLW described above. The following generally applicable definition is proposed:

Requires permanent isolation means concentrations of radionuclides that exceed the limits that are generally acceptable for near-surface land disposal as Class C waste, as defined in 10 CFR Part 61 as well as its supporting documentation and methodology.^{47,48}

Thus, the definition of a "long-lived" radionuclide is one that can occur in concentrations greater than its Class C limit, and knowledge of the concentrations of the most important long-lived radionuclides in the waste is needed for determining if the waste requires permanent isolation. The concentrations of radionuclides that define the permanent isolation boundary according to this definition are given in Table 2.18.

The waste classification system for defining HLW, TRU waste and equivalent, and LLW is depicted in Fig. 2.15 and may be summarized as follows:

1. HLW is waste in which (a) the power density exceeds 50 W/m^3 , or the external dose rate at

47. U.S. Nuclear Regulatory Commission, 10 CFR Pt. 61, 1982.

48. O. Oztunali and E. Roles, *Update of Part 61 Impact Analysis Methodology—Methodology Report, Vol. 1*, NUREG/CR-4370/V1, 1986.

Table 2.17. Selected radionuclide concentrations corresponding to highly radioactive boundary^a

Nuclide ^b	Boundary concentration (Ci/m ³)	Nuclide ^b	Boundary concentration (Ci/m ³)
^{14}C	2E5	$^{232}\text{U} + d$	2E2
^{63}Ni	5E5	^{238}Pu	2E3
$^{90}\text{Sr} + d$	7E3 ^c	^{239}Pu	2E3
$^{137}\text{Cs} + d$	5E3 ^c	^{240}Pu	2E3
^{151}Sm	4E5	^{241}Pu	2E6
$^{210}\text{Pb} + d$	1E3	^{241}Am	2E3
$^{226}\text{Ra} + d$	3E2	$^{241}\text{Am} + d$	1E3
$^{227}\text{Ac} + d$	2E2	^{243}Cm	1E3
$^{228}\text{Th} + d$	3E2	^{244}Cm	1E3
^{231}Pa	2E3	^{245}Cm	2E3

^aBoundary concentration for any radionuclide is based on a power density of 50 W/m^3 or an external dose-equivalent rate at a distance of 1 m from the waste of 100 rem/h (1 Sv/h), whichever is more restrictive; the highly radioactive boundary for wastes containing mixtures of radionuclides is determined from the boundary concentrations for each radionuclide using the sum-of-fractions rule or may be determined from direct measurements of power density and external dose rate.

^bNotation "+ d" denotes relatively short-lived daughter products assumed to be in secular equilibrium with the longer-lived parent.

^cValue corresponds to limit for near-surface land disposal as Class C waste (Source: U.S. Nuclear Regulatory Commission, 1982).

Table 2.18. Selected radionuclide concentrations corresponding to permanent isolation boundary*

Nuclide	Boundary concentration (Ci/m ³)	Nuclide	Boundary concentration (Ci/m ³)
¹⁴ C	8	²³² Th ^c	1E-2
¹⁴ C ^a	8E1	²³¹ Pa ^c	3E-2
⁵⁹ Ni ^b	2E2	²³² U ^c	5E-2
⁶³ Ni	7E2	²³³ U ^c	4E-1
⁶³ Ni ^b	7E3	²³⁴ U ^c	5E-1
⁹⁰ Sr	7E3	²³⁵ U	4E-1
⁹⁴ Nb ^b	2E-1	²³⁶ U ^c	6E-1
⁹⁹ Tc	3	²³⁸ U	5E-1
^{109m} Ag ^c	3E-2	²³⁷ Np	4E-2
¹²⁶ Sn ^c	1E-2	²³⁸ Pu	7
¹²⁹ I	8E-2	²³⁹ Pu	1E-1
¹³⁵ Cs	8E2	²⁴⁰ Pu	1E-1
¹³⁷ Cs	5E3	²⁴¹ Pu	5
^{180m} Pb ^c	2E2	²⁴² Pu	1E-1
²²⁶ Ra	3E-2	²⁴¹ Am	1E-1
²²⁷ Ac ^c	1	²⁴³ Am	7E-2
²²⁹ Th ^c	5E-2	²⁴³ Cm	8E1
²³⁰ Th ^c	6E-2	²⁴⁴ Cm	4E1

*Boundary concentration for any radionuclide is defined as the limit that is generally acceptable for near-surface land disposal as Class C waste (refs. 47 and 48). The permanent isolation boundary for wastes containing mixtures of radionuclides is determined from the boundary concentration for each radionuclide using the sum-of-fractions rule.

^aValue for radionuclide in activated metals only.

^bValue is not included explicitly in tables for 10 CFR Part 61 and supporting documentation and methodology (refs. 47 and 48) and, thus, is regarded as provisional.

- a distance of 1 m from the waste exceeds 100 rem/h (1 Sv/h) (i.e., radionuclide concentrations exceed the values in Table 2.16); and (b) concentrations of radionuclides exceed the limits that are generally acceptable for near-surface disposal as Class C waste in Table 2.18.
2. TRU waste and equivalent is waste in which (1) the power density is <50 W/m³ and the external dose rate at a distance of 1 m from the waste is <100 rem/h (1 Sv/h) (i.e., radionuclide concentrations are less than the values in Table 2.17) and (2) concentrations of radionuclides exceed the limits that are generally acceptable for near-surface disposal as Class C waste in Table 2.18.
 3. LLW is waste in which concentrations of radionuclides are less than the limits that are generally acceptable for near-surface disposal as Class C waste in Table 2.18, regardless of the levels of power density or external dose rate.
 4. In applying the boundary concentrations in Tables 2.17 and 2.18 to wastes that contain mixtures of radionuclides, the sum-of-fractions rule is used (i.e., the quantity to be calculated is the ratio of each radionuclide concentration to its corresponding boundary concentration, summed over all radionuclides, and the highly radioactive or permanent isolation boundary is exceeded if the appropriate sum of fractions exceeds unity).

It is not intended that all wastes defined according to the proposed classification system as HLW or TRU waste and equivalent (i.e., wastes with concentrations of radionuclides greater than the limits that are generally acceptable for near-surface disposal as Class C waste) necessarily require deep geologic repositories or equivalent. Some of these wastes may be candidates for greater confinement disposal (GCD), which we define as any technology that is more confining than near-surface disposal for Class C waste⁴⁷ but

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Fig. 2.15. Depiction of waste classification system. Radionuclide concentrations corresponding to boundaries defining high-level waste, transuranic waste and equivalent, and low-level waste are given in Tables 2.17 and 2.18.

less confining than a deep geologic repository or equivalent.

TRUPACT Support

The Chem Tech Division provided technical assistance to DOE in their activities related to establishing a transportation system to support the opening and operation of the Waste Isolation Pilot Plant (WIPP). This facility will accept transuranic wastes, and a transport package design does not currently exist which can be certified to meet the existing transport regulations. Two specific tasks were supported to assist DOE in obtaining approval for a packaging design.

At the instigation of the Chem Tech staff, the American National Standards Institute (ANSI) convened an ad hoc group under the auspices of the N14 Committee on the Packaging and Transportation of Radioactive Materials. The ad hoc group, which included a Chem Tech representative, was asked to consider several aspects of the pack-

aging and transportation of TRU waste and to provide recommendations about the requirements that are appropriate for the material.

The group reviewed background information related to two specific regulatory requirements pertaining to TRU waste: the prohibition against the use of filtered pressure equalization systems (filtered vents) and the requirement for use of double containment in the package design. The rationale for establishing these requirements and their applicability to the waste materials of interest were considered. The inhalation hazards of the plutonium waste forms and their chemical and physical forms within the waste were also taken into account. The group concluded that less rigorous packaging design criteria might be appropriate for contact-handled TRU waste meeting the WIPP Waste Acceptance Criteria. The hazards of the materials are bounded by the restrictions placed on them by the acceptance criteria; consequently, the full packaging requirements normally applicable to plutonium in powdered forms may be overly conservative. A recommendation was made to investigate establishment of a packaging category which would emplace design and performance requirements somewhere between the existing Type A and Type B packaging. The new packaging category would be restricted to materials which have their hazard limited by the imposition of upper bounds on dispersibility, respirability, and other hazardous characteristics.

In the formulation of an overall strategy for accomplishing an approved method to transport the TRU waste, DOE also requested that Chem Tech perform a technical review of the TRUPACT-I package design. Efforts were coordinated with participants brought in from other divisions, and an overall evaluation of the TRUPACT-I safety analysis report was prepared. This evaluation was supplied to DOE headquarters where it was used in revising the safety analysis report.

The second area in which Chem Tech assisted DOE relative to the TRU transport issue was the preparation of a draft petition for rulemaking to be submitted to the Department of Transportation (DOT). The petition was formulated on a basis which was consistent with the results of the ANSI group but built on specific design features and hazard limiting criteria. It requested that DOT allow the use of packaging designs which utilize filtered pressure equalization systems and without redundant containment systems. The packaging

designs would be restricted to transporting only contact-handled TRU waste of limited hazard (such as that complying with the WIPP Waste Acceptance Criteria). The petition was transmitted to DOE and was subsequently sent to DOT, where it is now undergoing review.

Transuranic Waste Management

The Defense TRU Waste Program for planning, integration, and technical development for TRU waste management is centered in the DOE's Albuquerque Operations Office and the Defense TRU Waste Lead Organization—the Joint Integration Office (JIO). The program's goal is to implement permanent disposal of these wastes from all defense waste locations and thus eliminate the need for interim storage. The WIPP, located in southeastern New Mexico, is to be used initially for retrievable emplacement tests and later, if those tests prove successful, for permanent disposal of defense TRU waste.

The TRU solid wastes at ORNL are classified by the radiation level at the exterior of the waste package. Hands-on operations may be utilized for contact-handled (CH) waste, which registers <200 mrem/h. Remote-handled (RH) waste, which measures >200 mrem/h, requires special (remote) handling for personal safety.

The CH-TRU waste program is more advanced than the RH-TRU waste program, reflecting the national plan to ship the CH wastes to the WIPP starting in late 1988, 5 years before transport of the RH-TRU wastes. The Operations Division at ORNL has primary responsibility for the preparation of certification documents, the design and construction of any packaging or repackaging facilities, and the ultimate shipping of TRU waste to the WIPP. Partly because it is a major producer of ORNL's TRU waste, Chem Tech has assisted in several efforts within the ORNL TRU waste program.

TRU Waste Certification. Certification of stored and newly generated CH-TRU waste began in FY 1986. A major factor in the certification of drummed CH-TRU waste at ORNL is the use of nondestructive assay (NDA) and nondestructive examination (NDE) techniques in the Waste Examination and Assay Facility (WEAF). Some of the drums will also be checked for integrity by an ultrasonic thickness probe. Chem Tech was instrumental in preparing specifications, evaluating bids,

and selecting a real-time radiography (RTR) unit for the NDE. This unit was tested on previously stored TRU waste and, after minor adjustments, turned over to the Operations Division for routine operations, where it has performed flawlessly.

In addition, all the preparation of necessary training and quality assurance (QA) documents needed for the certification of CH-TRU waste was coordinated by Chem Tech, with the ORNL program being accepted by the Waste Acceptance Criteria committee (WACC) and the final audit being completed in June 1986. If approved by ORNL certification procedures, CH-TRU waste is then considered acceptable for disposal at the WIPP. The programs for certifying other ORNL TRU waste have been prepared and sent to the WACC for review and acceptance. This review should be completed in FY 1987.

Certification of newly generated RH waste forms, excluding the container, began in January 1987. High-neutron-level counts will likely prevent the immediate shipment of the wastes to the WIPP, thus necessitating interim storage for 5 to 10 years to allow for decay of ^{252}Cf . Shipment of aged RH-TRU waste is scheduled to begin in January 1996, but this date will ultimately depend upon the construction of the Waste Handling Pilot Plant (WHPP). The WHPP has been proposed as the facility to process the ORNL RH waste presently contained in the concrete storage casks and transfer it into stainless steel drums or, if suitable, directly into RH canisters for shipment and emplacement in the WIPP. The WHPP, which will also have facilities for processing and packaging RH sludges, is expected to become operational toward the end of 1995. Shipment of certified TRU waste to the WIPP is scheduled to begin with CH-TRU waste in October 1988, with an expected completion date for final workoff in 2013.

Final Inventory Work-Off Plan. Chem Tech has also had responsibility for the preparation of ORNL's submission to the Defense TRU Waste Lead Organization's Final Inventory Work-Off Plan (IWOP). The Final IWOP addresses ORNL's overall strategy for retrieval, certification, and shipment of its stored and newly generated CH and RH-TRU wastes to the WIPP. Section 1 of the IWOP overviews ORNL's unique waste situation; Sect. 2 characterizes all the TRU wastes currently stored and projected to be stored at ORNL; Sect. 3 gives an overview of the methods

and facilities that will be needed to retrieve and certify the waste for shipment; Sect. 4 presents operational milestones and cost estimates, including costs for the long-term TRU waste management program; Sect. 5 defines the resource needs and tasks that need implementation; Sect. 6 lists the reporting requirements and public documentation required to support certification facilities and strategies; and Sect. 7, the Appendix, contains supporting information.

The main body of the report is Sect. 2, Waste Characterization. To provide the detail and waste characteristics that the JIO requested, it was necessary to extensively examine the TRU waste data in the ORNL Solid Waste Information Management Systems (SWIMS) as well as to make the first-time estimates of the wastes' characteristics.

In addition, the newly classified TRU waste, such as the Melton Valley sludges, had to be characterized. Calculations were made of the grams and curies of TRU nuclides contained in the Melton Valley tank sludge, the evaporator tank sludge, the gunite tank sludge, and the uranium oxide-containing cans stored at Building 3019. An isotopic distribution of elements in the RH-TRU waste planned to be shipped to WIPP was also prepared.

Contact-handled waste in storage at ORNL had traditionally been placed in stainless or mild steel drums (55-gal and 30-gal), while RH waste has been placed principally in concrete casks. Fifty-five-gallon stainless steel drums will probably be used to contain the RH-TRU sludges that resulted from ORNL's gunite tank sludge removal operations.

Remote-handled TRU waste presently totals 1264 m³, including 946 m³ of sludges. In addition, a projected volume of 1532 m³ of RH waste will be available before 2013, the projected end of the work-off period. After the WIPP begins handling RH-TRU waste (targeted for 1996), RH-TRU waste will be worked off at the rate of about 200 shipments per year. The CH-TRU waste presently totals 486 m³ and will increase to 1726 m³, requiring nine trips per year for a 25-year workoff. A conservative estimate for the cost of this total work-off effort for ORNL's TRU waste is \$153 million.

Because of the many changes in the existing and project inventory of RH-TRU waste at ORNL,

and because the IWOP contained the detail requested by JIO, it was decided by JIO that the 1986 IDB input should come from the waste-characterization tables of the current IWOP. This approach helps to ensure greater accuracy and consistency among the documents.

Because of the ongoing updating of TRU waste, changes have occurred with some of the previously listed waste volume and characterization data. Further, the integration of the TRU data from various data bases revealed many inconsistencies. Most of these inconsistencies have been corrected; however, future work will be necessary to ensure the reliability of the TRU entries, which, in turn, will result in an accurate submission of volumes and characteristics for ORNL's IDB annual inventory report.

Chem Tech was also asked to do a characterization of ORNL's special-case transuranic (SC-TRU) waste for the JIO to provide input into the development of a strategy document for SC-TRU waste. The SC-TRU waste is defined as any CH or RH-TRU waste identified by individual sites to be "difficult to certify" as is for emplacement in the WIPP. Such waste is characterized by unusual size, weight, high radiation level, or other constraints that might make WIPP certification impractical. Further, TRU waste that is to be sent to the WIPP is defined as defense waste, which in turn, is defined as "waste which results in whole or part from support of DOE atomic energy defense activities." Therefore, waste that was considered for the SC-TRU designation included (1) RH-TRU waste, (2) material that is encapsulated in stainless steel (SS) wells, (3) material from the Consolidated Edison Project (CEUSP), (4) Molten Salt Reactor fuel, and (5) some buried waste.

The characterization work for the JIO also provides input to a comprehensive plan related to the management of TRU waste at all DOE sites. The report provided more specific information of ORNL's inventory of SC waste than is presently contained in the Final IWOP.

Our RH-TRU waste is not considered SC-TRU waste because it will likely be certified at the proposed WHPP for emplacement in the WIPP. Historically, the material encapsulated in the SS wells had been regarded as TRU waste, but the current evaluation is that it is not SC-TRU because it is comprised of fuel elements. Also, this material originated chiefly from commercial sources. As

part of the RH demonstration plan, the CEUSP material is planned to be shipped to WIPP. The other forms for consideration are disqualified, by definition, from being SC-TRU waste. Hence, at present, ORNL has no SC-TRU-designated waste. Table 2.19 presents a summary and current evaluation of ORNL's previous candidates for SC-TRU consideration.

Table 2.19. Summary of SC-TRU candidates and current evaluation

Waste form	Past designation	Current designation
SS-well material	SC RH-TRU	<i>a</i>
66 Boxes of buried material	SC-TRU	TRU
Concrete casks	Both SC and RH	RH-TRU waste
MSRE fuel	Reactor fuel	Reactor fuel
CEUSP cans	SC RH-TRU	RH-TRU
Miscellaneous oxides	SC RH-TRU	Excess material

^aDependent upon acceptance by the Federal Waste Management Systems (FWMS); however, this material will not be considered for WIPP disposal.

Regulatory Analysis

Regulations by governmental agencies other than DOE often have considerable impact on DOE operations. Recently, the U.S. Environmental Protection Agency (EPA) has increased its authority to cover DOE installations, and the states have extended their influence also. Waste management, both at present and in the future, must be compatible with all regulations. Maintaining the expertise necessary to analyze the effects of "other agency" regulations on DOE operations is an important task in continuing the efficient operation of DOE facilities. In Chem Tech, a group of people trained in examining the impact of these regulations has been active for the past year. The overall effort on these activities has been multidivisional, coordinated by the Environmental Sciences Division, and supports the DOE Office of Environmental Guidance. The results are briefly discussed below.

The scope of the analyses on EPA's "Notices of Proposed Rulemaking (NPRM)" was very broad, as indicated by the list below.

- Land Disposal Restrictions: Double Liners (50 FR 10706)
- Groundwater Equations: Dilution/Attenuation (51 FR 21648)
- Identification and Listing of Hazardous Waste, Notification Requirements, and Reportable Quantity Adjustments: Toxicity Characteristics Leaching Procedure (51 FR 21647)
- Hazardous Waste Management System: Groundwater Monitoring (51 FR 26632)
- National Primary Drinking Water Regulations: Radionuclides (51 FR 34836)
- Emergency Planning and Community Right-to-Know Programs: Listing of Extremely Hazardous Materials (51 FR 41570)
- Standards for Owners and Operators of Miscellaneous Units Not Covered by Other EPA Regulations (51 FR 40726)

In-depth analysis of these proposals identified a number of areas within DOE's operation that would be significantly affected by these rules if they were promulgated. The information was transferred to DOE's Office of Environmental Guidance for use in responding formally to EPA on these issues.

Preparation of Environmental Analysis Documents. A new environmental analysis activity was initiated this year in providing technical support to the Integrated Analysis and Assessment Section (IAAS) of the Energy Division of ORNL. The primary emphasis of the IAAS is to assist the DOE, NRC, and other federal agencies with the preparation of environmental documents pursuant to complying with the National Environmental Policy Act (NEPA). The work is conducted in a matrix mode with interdisciplinary teams from other ORNL divisions, including Energy, Environmental Sciences, and Health and Safety Research. This initiative offers a bridge between the Chem Tech staff waste expertise and the regulatory programs.

Work in progress includes the preparation of a draft Action Description Memorandum (ADM) for the proposed Industrial Landfill III at the Y-12 Plant for mercury-contaminated wastes and a preliminary environmental analysis for the proposed decommissioning of the Wright-Patterson Air Force Base reactor. Chem Tech provides the team leader for these projects. Other activities this past year have included (1) a preliminary dose-estimate study of individual ORNL waste management sites

and ranking of the sites for remedial action based on dose; (2) the draft NEPA strategy document for renovating the DOE Fuel Materials Production Center (FMPC) located near Fernald, Ohio; and (3) the ADM for closing the Atomic Vapor Laser Isotope Separation Development Facility (AVLIS) at the Oak Ridge Gaseous Diffusion Plant (ORGDP).

Remedial Investigation/Feasibility Studies. The Oak Ridge National Laboratory contains many sites that have been identified as contributing known releases of radioactivity to the environment as well as many sites that are questionable contributors. Such sites must thus be addressed under Sect. 3004u of the 1976 Resource Conservation and Recovery Act (RCRA). Under the 3004u provisions, a remedial investigation/feasibility study (RI/FS) forms the basis for determining the extent of actions necessary for site stabilization and remedial action.

For the waste area grouping 4 (WAG4), which contains the inactive Solid Waste Storage Area 4 (SWSA-4), a data package was prepared which addresses such factors as the site description, history of known disposals and waste inventory, geology and soils, hydrology, and ecology. This document also contains pertinent engineering drawings, photographs, and references that will be useful in determining the remedial action that should be undertaken.⁴⁹ Similar reports for two additional WAGs, WAG3 and WAG5, will be completed in FY 1987.

SWSA-6 Site Characterization Support

Several related tasks in this effort have been undertaken to characterize the current active LLW disposal site at ORNL (Solid Waste Storage Area 6, or SWSA-6) with respect to current and anticipated future geochemical conditions and processes and to assist in estimating the current and projected future releases of radioactive and hazardous materials from waste already emplaced at the site. This information is needed to support performance assessment modeling of the potential releases from the site and will aid in the evaluation of remedial action options with respect to eventual site closure.

Release of Radionuclides from Neutron-Activated Wastes. Low-level radioactive waste sites at ORNL are being characterized to provide information to support the preparation of environmental statements and the analysis of site closure options.

The objective of this task is to model the near-field radioactivity release rates for some of the high-activity LLW currently emplaced in SWSA-6.

Reactor components are discharged from the High Flux Isotope Reactor (HFIR) and emplaced in SWSA-6 at ORNL. The components are radioactive due to neutron activation of various stable isotopes. Emplacement involves disposal in lined and unlined auger holes in soil located above the water table. The radionuclide inventory of disposed components, as of June 1, 1986, was calculated. Information on the composition and weight of the components as well as reasonable assumptions for the neutron flux during use, the time of neutron exposure, and radioactive decay after discharge were employed in the inventory calculation. The near-field (in the immediate vicinity of the emplaced wastes) release rates of ¹⁵²Eu, ¹⁵⁴Eu, and ¹⁵⁵Eu from control plates and cylinders were calculated for 50 years after emplacement. The release rates of the europium isotopes were uncertain because of the lack of information for a number of the calculational parameters. Two release-rate-limiting models were considered (a saturation-limited model and a diffusion-limited model), and ranges of reasonable values were assumed for the time to failure of the auger-hole liner and aluminum cladding and for the europium solubility in SWSA-6 groundwater. The bounding europium radionuclide and near-field release rates peaked at ~1.3 Ci/year total for ¹⁵²Eu, ¹⁵⁴Eu, and ¹⁵⁵Eu in 1987 for the lower bound and at ~420 Ci/year in 1992 for the upper bound. Better input information allowed calculation of the radioactivity releases from the other components with more confidence. The near-field release rates of ⁵⁵Fe, ⁵⁹Ni, ⁶⁰Co, and ⁶³Ni from stainless steel and cobalt alloy components, as well as of ¹⁰Be, ⁴¹Ca, and ⁵⁵Fe from beryllium reflectors, were calculated for the next 100 years, assuming bulk waste corrosion to be the release-rate-limiting step. For the stainless steel and cobalt alloy components, the current (1986) total radionuclide release rate was calculated to be ~0.07 Ci/year, which decreases by 2010 to a steady release of ~0.003 Ci/year due primarily to ⁶³Ni. Under the most conservative assumptions for the reflectors, the current (1986)

49. E. C. Davis and R. R. Shoun, *Environmental Data Package for ORNL Solid Waste Storage Area Four, the Adjacent Intermediate-Level Liquid Waste Transfer Line, and the Liquid Waste Pilot Pit Area*, ORNL/TM-10155, September 1986.

total radionuclide release rate was calculated to be $\sim 1.2 \times 10^{-4}$ Ci/year, decreasing by 1992 to a steady release of $\sim 1.5 \times 10^{-4}$ Ci/year due primarily to ^{41}Ca . The dominant uncertainties in the calculation of the inventory and release rates, as well as the research and development work that would be needed to reduce these uncertainties, have been considered.

Leachability of Low-Level Wastes. A series of tests was completed to explore the leachability of radionuclides from typical LLW currently being emplaced in trenches in SWSA-6. Leaching of both radioactive contaminants and RCRA hazardous material contaminants from representative LLW generated at ORNL was investigated using two different leaching methodologies: (1) the current U.S. Environmental Protection Agency (EPA) Toxic Waste Extraction Procedure (EP) and (2) a proposed EPA Toxicity Characteristic Leach Procedure (TCLP). Cesium, both as ^{137}Cs and ^{134}Cs , was present in many of the waste samples. The average fraction leached for cesium was only ~ 0.4 . Since much of the cesium handled at ORNL is in the form of CsCl , the limited leaching observed was surprising. Cobalt, as ^{60}Co , was present in many samples; the fraction leached ranged from ~ 0.2 to 0.7 for various samples. Strontium, as ^{90}Sr , was present in all but one waste sample. Strontium was readily leached from most samples; this result seemed reasonable because much of the strontium handled at ORNL is in the form of SrCl_2 . Selenium, present as ^{75}Se , was identified in one-half of the samples tested. This observation was surprising, because selenium is not currently listed as a radioactive waste component in ORNL LLW. The selenium fraction leached was only ~ 0.2 . Chromium, cadmium, and lead were the only inorganic elements identified in a few waste samples at sufficient concentrations to be of potential environmental concern. In no case, however, did the leach extract exceed the EPA toxicity characteristic limit. For a number of the wastes tested, the proposed TCLP leach methodology was more effective in extracting contaminants than was the current EP method.

Analysis of SWSA-6 Trench Leachates and Groundwaters. An ongoing activity involves the sampling and analysis of trench leachates and groundwaters of the SWSA-6 site. The purpose of this work is to provide information descriptive of the current radiological and chemical contamination of the site as well as to help define the site

source term for the calculation of future performance. Three trench leachate samples and two groundwater samples have been collected during this report period. A total of 14 samples were planned; however, unusually low precipitation during this report period has resulted in many dry trenches and has caused delays in sampling. All samples collected to date contain appreciable quantities of beta-emitting radionuclides (primarily ^3H and ^{90}Sr). A number of EPA priority pollutant organic compounds were detected, including naphthalene and toluene, which are present at appreciable concentrations. All samples exceeded the EPA Interim Primary Drinking Water Standards for alpha- and beta-emitting radionuclides. Two samples contained concentrations of organics (chloroform, methylene chloride, and naphthalene) in excess of the State of Tennessee Guidelines for Superfund Sites.

2.9 SYSTEMS ANALYSIS FOR THE LOW-LEVEL WASTE DISPOSAL DEVELOPMENT AND DEMONSTRATION PROGRAM

The three-plant Oak Ridge Complex produces a large quantity of various wastes ranging from high-level radioactive to nonradioactive hazardous types. The disposal and discharge of these wastes are regulated by many state and federal laws, and several new programs have been initiated in order to comply with these laws. One of these is the Low-Level Waste Disposal Development and Demonstration (LLWDDD) Program, which has as its objective to provide a technical focal point for the development and deployment of new, acceptable low-level waste (LLW) management facilities on the Oak Ridge Reservation in a manner which will ensure continuation of normal activities by the local DOE facilities. To meet this objective, the program must first develop an in-depth understanding of current operations and then devise a plan for integrating the operations using the latest technology while maintaining the lowest costs compatible with environmentally acceptable practice. This is being accomplished by a systems analysis subtask which (1) determines the current state of waste management on the Oak Ridge Reservation, (2) tabulates and organizes the information into a usable data base to support future activities, (3) assists in the selection of technologies

to demonstrate, and (4) follows facility design and construction through to completion of integrated operations while constantly updating technology and cost information. In other words, this subtask provides the foundation of knowledge upon which future decisions will depend.

The systems analysis subtask consists of three parts: (1) technology/costing evaluations; (2) integration of current activities into a single dynamic flowsheet (i.e., a flowsheet that will be continually changing), supported by a relational data base that links all available information; and (3) process and flowsheet simulations that allow examination of various technology configurations before expenditures for construction are committed. During this first year, activity has been concentrated in the first two areas, which are discussed below.

Relational Data Base and Dynamic Flowsheet Development

The development of the data base involves incorporating information from the entire Oak Ridge Reservation. This information, frequently scarce in particular areas, is derived from varied sources and exists in different formats. It must be integrated into a single, logical format that can be easily searched by various computer programs. The data base, which is still under development, utilizes the VAX relational data-base product RdB® and is being developed on the Chemical Technology Research Microvax II. Some of the input received so far is discussed below.

The Liquid Source Characterization Data Base for X-10 (SCDX), which contains detailed information on LLW sources at the X-10 site, has been obtained. This information, which consists of flow rate, analytical, and radiological data, along with source descriptions and details of the buildings, was initially created in the dBASE III Plus® format. However, most of it has already been incorporated into the relational (RdB®) data-base structure.

Data derived from the solid waste information management system (SWIMS) data base for waste managed at the X-10 site have been obtained in the form of a KMAN® data file. These data include waste quantities, estimates of radionuclide content, and other pertinent information. Activities are under way to convert the data from KMAN® format to ASCII format for inclusion in the RdB® data base being developed.

Data from a limited waste characterization study by Martin Marietta Energy Systems, Inc., Engineering, for the three plants on the Oak Ridge Reservation have been obtained. Included in this Lotus 1-2-3® data base is information on current-year and projected waste amounts, as well as descriptions and characteristics for some general waste stream categories which are currently expected to be stored at K-25. These data will be converted to RdB® format for future use.

Each of these data bases, along with other data currently in hand or to be obtained in the future, will become a contributor to a larger, integrated data base of waste stream information for all the Oak Ridge Operations (ORO) facilities. This expanded data base is to be used by the Oak Ridge Model, which is under development, to aid in the evaluation of LLW management alternatives within DOE. The results will serve as a model for all DOE facilities.

The flowsheet model is a tool for use in scoping studies designed to determine and prioritize needs for waste characterization studies and technology demonstrations. As it evolves, it will allow planners to estimate more accurately the impacts of changes in process operations and operational procedures on the waste quantities generated.

The three-plant Oak Ridge Model is represented by three block flow diagrams (one for each plant). Each of these diagrams is divided by a seven-column format, with each column representing one of the steps along the waste flow path: (1) generation, (2) staging (less than 90-d storage), (3) treatment, (4) storage (permitted and greater than 90 d), (5) disposal, (6) discharge, and (7) off-site transfer. The facilities at the three plants are grouped in a manner which is conducive to locating information on a specific step of the flow path (e.g., types of treatment). The generated waste streams, solid and liquid, are grouped according to the type of waste generated. The various types of waste include (1) low level, (2) hazardous, (3) mixed, (4) TRU, (5) sanitary, (6) process, and (7) nonradioactive nonhazardous. As previously stated, this waste grouping allows us to concentrate on the streams that contain the waste of interest. For completeness, future waste streams, treatment facilities, storage locations, and disposal sites are also indicated on the drawings to the extent that future planning allows.

In the future, these flowsheets will be converted into a computer-simulated model, which can easily

be kept updated as changes in waste treatment operations occur. Finally, when the simulated flowsheet model is combined with the waste-stream data base that is also being developed, we will be able to perform computer simulations of how various waste treatment operations will affect the amount of waste that has to be disposed of and determine how the cost of this treatment compares with the amount saved in disposal costs.

Status of Modeling Effort

Process simulation, systems modeling, and economic evaluation comprise a major element contributing to the Waste Management Technology Center's LLWDDD program. The purpose of this effort is to support the analysis of waste management options and the evaluation, development, and implementation of technology demonstrations. Analysis at two levels of detail is envisioned. A simplified overall waste system model is being developed to aid strategy formulation, definition of requirements, system economic evaluation, etc., as discussed in the previous section. Detailed simulations to assess specific processes and treatment technologies will also be developed in the future.

The integrated systems model is being constructed based on the three-plant flowsheets and the quantity and characterization information in the data base described above. This method will provide a means for studying the interactions in the overall waste system and a framework for economic analysis and sensitivity studies. The model, which is being programmed in FORTRAN, will track each major waste stream from source through disposal. Attributes such as flow rates, radioactivity, compactibility, etc., will be associated with each stream; and the costs associated with

segregation, storage, treatment, packaging, and disposal will be included. Simple physical treatments (e.g., compaction and segregation) can be incorporated easily into the integrated systems model; however, other chemical and separation processes will require more rigorous modeling.

Detailed process simulations will be undertaken, where appropriate, to evaluate process alternatives, improve operations, and assess economics. Areas that may benefit from rigorous simulation include those processes which produce solid waste streams and nonmechanical volume reduction processes such as incineration. The particular approach that is taken will depend on the scope and level of detail of the information that is desired about the process, whether steady-state or dynamic simulations are required, and so forth. General-purpose codes, such as ASPEN* and ACSL,† will be used wherever feasible. However, other general process simulators as well as process specific codes will continue to be evaluated with regard to their applicability to the diverse requirements of this task.

The first application will be to use a preliminary FORTRAN version to examine segregation of wastes and the economics associated with the various alternatives available, which depend on regulations and class delineations. Some results from this model are expected in the first quarter of 1987.

*ASPEN (Advanced Simulator for Process Engineering) is a general-purpose steady-state process code simulator developed at the Massachusetts Institute of Technology.

†ACSL (Advanced Continuous Simulation Language) is a commercially developed language designed for modeling and evaluating the performance of continuous systems described by time-dependent nonlinear differential equations.

3. ENVIRONMENTAL CONTROL TECHNOLOGY

Environmental control technology (ECT) continues to be an expanding area in Chem Tech. The current staffing for this activity is seven engineers and three technicians on a full-time-equivalent basis. Work for external sponsors such as the Tennessee Valley Authority, the Electric Power Research Institute, the U.S. Department of Defense, and other DOE/Oak Ridge Operations facilities has increased significantly during this report period. The largest fraction of work has, however, continued to be the support to organizations within ORNL. A major objective for the group is to become recognized within the larger DOE system as the center for R&D for ECT, as has already taken place with regard to work within the ORNL complex. Progress related to projects conducted within the Oak Ridge complex is summarized below.

3.1 SUPPORT TO THE OAK RIDGE COMPLEX

Chem Tech's Environmental Control Technology Group supplies experimental development and engineering analysis support to the Environmental Restoration and Facilities Upgrade (ERFU) Program and the Operations Division in several areas. The current primary area of activity is the liquid waste processing system at ORNL. Present efforts are subdivided into four general areas: (1) nonradiological wastes, (2) Process Waste Treatment Plant support, (3) low-level waste treatment, and (4) other studies. Projects within these areas are discussed individually below.

Nonradiological Wastes

Treatability Studies in Support of the Nonradiological Wastewater Treatment Plant. The Nonradiological Wastewater Treatment Plant (NRWTP) will treat nonradiological wastewaters generated at ORNL to reduce pollutant levels to those acceptable under restrictions imposed by the effluent lim-

its of best available technology (BAT) regulations of the U.S. Environmental Protection Agency (EPA), according to the goals established by the Clean Water Act.¹ The effluent standards that are listed in the NPDES permit² are shown in Table 3.1.

Table 3.1. NPDES effluent limits for the Nonradiological Wastewater Treatment Plant

Parameter	Monthly average (mg/L)	Daily maximum (mg/L)
Total suspended solids	31	60
Oil and grease	10	15
Total toxic organics		2.13
Cyanide	0.65	1.20
Cadmium	0.26	0.69
Chromium	1.71	2.77
Copper	2.07	3.38
Lead	0.43	0.69
Nickel	2.38	3.98
Silver	0.24	0.43
Zinc	1.48	2.61

Source: National Pollutant Discharge Elimination System Permit No. TN0002941, Pt. I, pp. I-38-41.

The influent to the NRWTP will come from several sources and will typically contain occasional surges of the heavy metals (milligram-per-liter levels or less), lasting up to a few hours, and a wide variety of criteria organics at the microgram-per-liter (parts-per-billion) level. The handling of this wastewater is not a typical treatment problem because of its dilute nature, widely varying composition, and high flow rate—0.022 m³/s (350 gpm) nominal up to 0.035 m³/s (560 gpm) maximum.

1. *Environmental Statutes 1986 Edition*, pp. 231-366, Government Institutes, Inc., Rockville, Md., April 1986.

2. National Pollutant Discharge Elimination System Permit No. TN0002941, Pt. I, pp. I-38-I-41.

To resolve many of the uncertainties facing the NRWTP, ORNL engaged in a three-phase treatability study. Initial scouting work was done in which granular activated-carbon (GAC) adsorption and reverse osmosis (RO) were performed on two of the major feed streams to the NRWTP.³ Phase I of the main study consisted of batch simulation of the proposed NRWTP flowsheet in the laboratory; results revealed no major problems with the proposed flowsheet. Phase II consisted of more detailed parametric studies of the flowsheet processes at a bench-scale level in the laboratory. The following bench-scale treatability studies were performed: titration curves; jar tests to determine the optimum precipitation chemistry; polymer selection; column settling tests; filterability tests; air-stripping experiments; carbon isotherm studies; and heavy-metal-selective, ion-exchange resin tests.

The Phase II results were used to guide the planning and design of the Phase III study, which consisted of flowsheet simulation on a continuous basis using a mini-pilot-plant (MPP) facility. This facility, which is contained within two connected semi-trailer vans and an analytical trailer, is shown schematically in Fig. 3.1. The purpose of the MPP was to simulate the treatment the ORNL nonradiological wastewaters will receive in the NRWTP.

In the MPP, all wastewaters with the exception of those not containing heavy metals (i.e., Cd, Cr, Cu, Fe, Pb, Hg, Ni, Ag, and Zn) flowed through an API-type separator. The criteria metals (except

3. J. F. Walker and C. H. Brown, Jr., *Scouting Treatability Studies Performed in Support of the Nonradiological Wastewater Treatment Facility Feasibility Study*, ORNL/TM-9667, April 1986.

ORNL DWG 85-1142R

NOTE: TRAILERS JOIN VESTIBULE AT A--A AND B--B LINES SHOWN ABOVE.

Fig. 3.1. Layout of mini-pilot plant.

for mercury) were removed in the reactor/clarifier (R/C) as hydroxide precipitates using either calcium hydroxide (lime) and sodium carbonate (soda ash) or sodium hydroxide (caustic). Overflow from the precipitator was then treated by dual-media (sand and anthracite) filters and adjusted to a pH of 7. Wastewaters that had been pretreated by ion exchange for radionuclide removal bypassed the first four unit operations (phase separation, heavy-metal precipitation, filtration, and pH adjustment) and were combined with the other wastewaters prior to being air-stripped. Volatile organics were removed by air-stripping in a packed tower, while heavy (nonvolatile) organics and mercury were removed in three serially connected GAC columns. An ion-exchange column was operated in parallel with the R/C in order to evaluate the long-term performance of a chelating, heavy-metal-selective resin.

In general, results of the MPP studies correlated well with those of the bench-scale treatability studies. Operation of the MPP demonstrated that cleanup to levels approaching federal drinking water standards can be achieved 98% of the time and that control of heavy metals at levels <0.1 ppm at all times is not an unreasonable goal for the ORNL nonradiological wastewaters when the selected unit processes⁴ are used.

Development of a Continuous, Automated Monitor for Wastewater Metals. ORNL generates process wastewaters with highly variable characteristics because of the diverse nature of activities contributing wastes. The concentration levels of several trace metals in the wastewater must be determined prior to wastewater treatment and final discharge through an NPDES discharge point. To gain reasonable knowledge of average and peak amounts of specific trace metals in the combined drainage system, continuous monitoring is essential. Toward this end, a relatively inexpensive, sensitive, remote metals analyzer system was developed.

In this system, all metals of concern (Cd, Cr, Cu, Ni, Pb, and Zn) can be analyzed in near real time at the parts-per-billion level by polarographic means. Two analytical methods are used by this system: (1) differential-pulse polarography (DPP) for the determination of chromium and nickel and (2) differential-pulse anodic stripping voltammetry (DPASV) for the determination of Zn, Cd, Pb, and Cu.

The basic equipment comprising the analyzer is available commercially. At the heart of the system (see Fig. 3.2) are two Princeton Applied Research (PARC) Model 309 Automatic Voltammetric Electrodes. The system provides for automatic and cyclic withdrawal of a sample from a waste stream side loop, with programmed analysis and discard of the sample. A PARC Model 384B Polarographic Analyzer, a programmable potentiostat, controls the electrodes and makes the current-voltage measurements. The 384B has an integral microcomputer for data processing and is interfaced with a personal computer for timing, data acquisition, and data storage. The sample is extracted from a circulated waste side stream, mixed with an electrolyte solution, and delivered to the electrode by a peristaltic pump. The cyclic pump action is activated by control circuits installed in the electrode modules.

Data from analysis of a variable-composition process waste stream at ORNL indicate the following sensitivities in on-line operation:

Metal	Sensitivity ($\mu\text{g/L}$)
Cd	1
Cr	30
Cu	30
Ni	30
Pb	5
Zn	20

Although the present system (see Fig. 3.3) was developed for monitoring metals in relatively clean process wastewaters, the analytical methods can be adapted for determination of other metals, anions, and electroactive organics without additional equipment. Pretreatment schemes are also foreseeable for the application of this method for streams containing interferences, such as complexing agents, surfactants, and certain organics.

Process Waste Treatment Plant Support Operations

Process aqueous wastes generated at ORNL are currently collected in an equalization basin and processed at the Process Waste Treatment Plant

4. J. M. Begovich, C. H. Brown, Jr., J. F. Villiers-Fisher, and V. L. Fowler, *Treatability Studies in Support of the Nonradiological Wastewater Treatment Project*, ORNL/TM-10046, July 1986.

Fig. 3.2. Schematic diagram of the on-line polarographic metals analyzer.

Fig. 3.3. Equipment associated with the on-line polarographic metals analyzer.

(PWTP), located in Building 3544, to remove radioactive cations (primarily ^{90}Sr and ^{137}Cs). At the beginning of the report period, the wastewater was treated by a strong-acid cation-exchange resin that was regenerated with HNO_3 . The regenerant solution was evaporated, neutralized with caustic, and sent to the liquid low-level radioactive waste (LLLW) facilities for final disposal. Since discontinuance of use of the hydrofracture process (the former method of ultimate disposal of LLLW), the Operations Division requested that Chem Tech evaluate the PWTP to improve efficiency and reduce waste generation. The primary goals of this project have been to (1) implement process improvements at the PWTP to immediately reduce liquid waste generation, (2) improve process reliability, and (3) provide a basis for design of an upgraded facility.

Process Improvements at the PWTP. A number of tests were performed in an effort to improve the efficiency of the existing PWTP. Some of the resulting improvements have been implemented at the PWTP to reduce waste generation. The results are summarized below.

1. A clarifier, previously used at the PWTP in the scavenging-precipitation ion-exchange process, has been installed upstream of the ion-exchange columns.⁵ A caustic softening process is being used to remove calcium and magnesium, which compete with the radionuclides for sites on the resin. The precipitate, which contains ~70% of the ^{90}Sr , 20% of the ^{137}Cs , and a significant fraction of the ^{60}Co and ^{152}Eu when present, is being dewatered by a plate-and-frame filter press and temporarily stored in 55-gal drums at the Oak Ridge Gaseous Diffusion Plant. This upgrade has reduced liquid waste generation by 85% and total waste generation by 35%.
2. Prior to installation of the clarifier, the ion-exchange columns were often regenerated so frequently that the evaporator could not accommodate the total regenerant flow. The regenerant was neutralized with sodium hydroxide and pumped directly to the LLLW system without volume reduction. To eliminate this problem, the evaporator capacity was raised by increasing the steam pressure and installing an additional condenser in parallel with the existing one. Since installation of the clarifier, the upgraded evaporator has adequately handled all regenerant wastes.

3. A study was undertaken to determine the feasibility of steam-stripping the evaporator condensate rather than neutralizing it with caustic prior to pumping it to the LLLW system. Implementation of the stripping and distillation processes could lead to a 50% reduction in waste salt production.
4. Bench-scale studies show that the volume of regenerant solution sent to the LLLW system can be reduced by a factor of ~2.7 by extending the evaporation cycle until the boiling point reaches 135°C . Tests indicate that deeper evaporation will be impractical unless the waste regenerant contains a negligible amount of sodium nitrate. Bench tests indicate that steam-stripping will remove the excess acid remaining after distillation to 135°C . Since solids deposition is possible at temperatures below 50°C , all process lines would need to be heated. This constraint may make it necessary to process the waste on-site rather than through the LLLW system. The addition of quicklime solidifies the molten salts, neutralizes the excess acid, and ties up the water and weakly bounded hydrates.
5. Algae proliferation in the equalization basin has caused operational problems at the PWTP (i.e., inhibits sludge filtration and causes pressure buildup in resin columns). Regular addition of 0.5 to 3 ppm copper as Cutrine-Plus,^{*} a copper-based algicide, to the basin has adequately controlled biogrowth. A gasoline-powered sprayer system has been assembled on a small, low-bed trailer for algicide addition to the basin.

Evaluation of Instrumentation for the PWTP.

The evaluation of a facility upgrade instrumentation and control package for the PWTP has been completed. The purpose for this evaluation was to determine if certain instrumentation and control systems could be used to improve the operating efficiency of the PWTP. Evaluations were conducted on the following instruments and related equipment:

1. on-line total hardness monitors for determination of ion-exchange-column total hardness breakthrough,

5. S. M. Robinson and J. M. Begovich, *Treatment Studies at the Process Waste Treatment Plant at Oak Ridge National Laboratory*, ORNL/TM-10352, to be published.

^{*}Cutrine-Plus is a trademark of Applied Biochemists, Inc.

2. flame photometer for potassium analyses,
3. programmable multipoint recorder,
4. microprocessor-based pH analyzer/controller for final pH adjustment of the PWTP effluent,
5. various pH monitoring systems,
6. metering pumps used for pH adjustment of final effluent,
7. on-line total organic carbon analyzer for determination of organics in PWTP effluent,
8. on-line gamma counter with flow-through sodium iodide detection system for ^{137}Cs detection in PWTP effluent,
9. automatic polymer mixing and addition unit for adding flocculant to the PWTP influent stream, and
10. automatic ferric sulfate feed system for addition of iron to aid the precipitation process.

Recommendations were made to the Operations Division regarding the relative ease of operation, maintenance and labor requirements, and suitability for application for each of the above systems. Several of the instrumentation systems did not live up to their respective expectations. Some systems became obsolete as the evaluation period progressed because of changes in PWTP operation. The systems that survived the evaluation consisted mainly of the programmable multipoint recorder, the pH control systems, the automatic polymer addition unit, and the automatic ferric sulfate addition system. These units alone save many man-hours of labor and have greatly improved the operating efficiency of the PWTP.

Ion-Exchange Pilot Facility. In the near future, the PWTP will be required to meet more stringent effluent limits, whether the discharge is treated further in the NRWTP or the effluent is piped directly to White Oak Creek. Several modifications to the system have been proposed. The efficacy of these changes can be judged only through testing.

An engineering-scale facility (ESF) that performs the same function as the current configuration of the PWTP (i.e., cation exchange using HCR-S resin⁶) has been designed, constructed, installed, and operated. A series of experiments that model the present operating parameters of the PWTP has been completed. Specific objectives of these experiments were to improve resin regeneration and to enhance detection of ^{137}Cs in the plant effluent. The present procedure for determining the

point at which an ion-exchange column in the PWTP is taken off-line is by effluent analysis for total hardness (TH). A TH value of 2 mg/L (as CaCO_3) has been used to indicate the saturation of the resin for ^{137}Cs and/or ^{90}Sr . This simple criterion has historically been used because of the time required for radioisotope analysis. Because of the order of cation selectivity of HCR-S ion-exchange resin, an effort was made to determine the feasibility of using the potassium content (analysis by flame photometry) of the effluent to predict the cesium breakpoint.

As can be seen by the normalized data (i.e., effluent concentration divided by feed concentration vs throughput expressed as the number of bed volumes of wastewater processed) presented in Fig. 3.4, potassium in the effluent was first detected (0.1 mg/L) at 250 bed volumes (BV). Cesium content at this time was 67 Bq/L with a TH content of <1 mg/L. A TH break of 2 mg/L occurred at 680 BV, with the effluent containing 728 Bq/L of ^{137}Cs . Natural strontium was first detected in the effluent (0.005 mg/L) at 700 BV. The TH at this point was 6.3 mg/L. Based on the above analyses, TH is not a good indicator for the cesium break; and potassium may or may not be a reliable indicator, depending on future ^{137}Cs release limits. For control of the release of strontium, TH appears to be a reliable indicator, but radionuclide assays for ^{90}Sr are needed to verify this.

The ESF will be used in the future to determine the effects of using raw or softened wastewater on the radionuclide sorption capacities of various organic ion-exchange resins and inorganic zeolites. The sorbents tested will be those deemed most promising in our flowsheet development studies.

Flowsheet Analysis for Upgrade of the PWTP. Improved chemical precipitation and ion-exchange methods are being developed at ORNL in an effort to reduce waste generation at the PWTP. A wide variety of screening tests was performed on potential precipitation techniques and ion-exchange materials on a laboratory scale. The data were used to develop potential flowsheets for use at the PWTP; these flowsheets have been modeled to

6. V. L. Fowler, J. M. Begovich, J. F. Villiers-Fisher, and J. D. Hewitt, *Design and Operation of an Ion Exchange Pilot Facility for Cesium and Strontium Sorption*, ORNL/TM-10353, to be published.

Fig. 3.4. Breakthrough curves for Sr, Cs, Na, K, total hardness, and Eu as measured in the ion-exchange pilot facility.

determine the operating conditions and waste generation at full scale. Each flowsheet was evaluated using future-value economic analyses and performance ratings to determine the most effective processes to be considered for further development.

Four types of precipitation processes (needed to remove calcium and magnesium ions that compete with the radionuclides for adsorption sites on the sorbents) and 17 ion-exchange materials (including organic weak-acid and strong-acid cation resins and synthetic and natural zeolites) were evaluated in laboratory-scale screening tests using actual wastewater. These tests resulted in the selection of two caustic-based softening processes to be used in

conjunction with ion-exchange resins in proposed process flowsheets. Results from the column tests indicated that most of the zeolites have high sorption capacities for cesium from unsoftened process water. None of the materials tested have extremely high sorption capacity for strontium in untreated wastewater; chabazite zeolites were the best. Several materials, for example, HCR-S strong-acid resin, IRC-84 weak-acid resin, and clinoptilolite zeolites, are good sorbents for strontium when the feed has been pretreated by either of the softening processes.

Five process flowsheets containing the above-mentioned materials were developed for further

consideration. The laboratory-scale data were fitted with models developed by Thomas⁷ and Wallace⁸ and used to predict the performance of the full-scale ion-exchange columns. Cost comparisons for the flowsheets were made by calculating the future value of each flowsheet based on a 15-year plant life. Costs included capital costs for new equipment, plant operating costs, and waste disposal costs. Since many factors other than cost will influence flowsheet selection, the flowsheets were evaluated by numerically rating the processes on costs, plant operability, process flexibility and simplicity, stage of development, waste reduction, environmental and occupational safety, postprocessing requirements, and ultimate disposal requirements.

Both the economic analyses and flowsheet evaluations⁹ lead to the same conclusion: The flowsheet in which chabazite columns operated in series is used is the most attractive process for use at the PWTP. The next-best alternatives are flowsheets that consist of (1) a precipitator followed by clinoptilolite columns operated in series; (2) chabazite columns to remove ¹³⁷Cs, a precipitator, and IRC-84 resin columns to remove ⁹⁰Sr; and (3) chabazite columns to remove ¹³⁷Cs, followed by a continuous countercurrent ion-exchange column to remove ⁹⁰Sr. The existing process was rated the lowest by a significant amount. The proposed flowsheets have the potential to reduce the total waste generation by 70% and total PWTP operating costs by 60% compared with the original ion-exchange flowsheet.

Pilot- and Full-Scale Testing of Zeolite Columns at the PWTP. In December 1985 and January 1986, a significant spike occurred in the concentrations of ⁹⁰Sr and ¹³⁷Cs entering the process wastewater collection basin feeding the PWTP. Additional treatment equipment was needed temporarily to supplement the PWTP. Thus a decision was made to install skid-mounted equipment, leased on an emergency basis, and to perform a full-scale test (based on our flowsheet analyses studies for upgrading the PWTP) using columns filled with Linde Ionsiv IE-95 synthetic zeolites (manufactured by Union Carbide Corporation).

During the three weeks the test was conducted, the feed to the plant ranged from 2,400 to 7,100 Bq/L in ⁹⁰Sr concentration and from 320 to 720 Bq/L in ¹³⁷Cs concentration. Effluent concentrations indicated that the ¹³⁷Cs did not break

through either of the columns. Log-normal plots of the breakthrough curves for ⁹⁰Sr from both columns are shown in Fig. 3.5. These data confirmed the ⁹⁰Sr and ¹³⁷Cs removal efficiencies that had been obtained in the small-column tests. Modeling¹⁰ of the ⁹⁰Sr experimental data resulted in the curves shown on the figure.

Further testing on 0.1-plant-scale equipment has been continued to (1) evaluate other promising sorbents, (2) develop operating techniques for a series of four columns, and (3) develop the back end of the process, consisting of treatment and packaging of the spent sorbent. Currently in progress is the evaluation of a natural chabazite type of zeolite, TSM-300, supplied by Tenneco Specialty Minerals at less than 10% of the cost of Ionsiv IE-95. The performance of the TSM-300 has been much better (after one cycle) than that of the IE-95. Because of the high pressure drop across the first column, the system was shut down after processing 8,200 BV of wastewater. Additional cycles will be performed after a new sand filter has been installed.

Zeolite Posttreatment Studies. Heat treatment has been proposed as a potential method of preparing loaded zeolites for ultimate disposal. A zeolite is a molecular sieve-type material with a distinctive crystalline structure. When zeolites are heated to near 1,000°C, their crystal structures are destroyed and their pores collapse. This should result in shrinkage of the particles to approximately half their original volume. Theoretically, their leachability should be reduced because diffusion of

7. N. K. Hiester, T. Vermeulen, and G. Klein, *Chemical Engineers' Handbook*, 4th ed., ed. J. J. Perry et al., McGraw-Hill, New York, 1963.

8. E. D. Collins, D. O. Campbell, L. J. King, J. B. Knauer, and R. M. Wallace, "Development of the Flowsheet Used for Decontaminating High-Activity-Level Water," pp. 212-27 in *The Three Mile Island Accident: Diagnosis and Prognosis*, ed. L. M. Toth, A. P. Malinauskas, G. R. Eidam, and H. M. Burton, ACS Symposium Series 293, American Chemical Society, Washington, D.C., 1986.

9. S. M. Robinson, ORNL, personal communication to J. B. Berry, ORNL, Mar. 5, 1987.

10. E. D. Collins, J. M. Begovich, C. H. Brown, Jr., D. O. Campbell, L. C. Lasher, M. I. Morris, S. M. Robinson, and C. B. Scott, "An Improved Ion Exchange Method for Treatment of Slightly Contaminated Waste Waters," presented at the American Nuclear Society International Meeting on Low-, Intermediate-, and High-Level Waste Management and Decontamination and Decommissioning, Niagara Falls, N. Y., Sept. 14-18, 1986; proceedings to be published in 1987.

feed pH of 12. Rejection decreases to 80% at a feed pH of 14.

Batch filtration of MVST waste was accomplished via Millipore filters of 0.45- μ m nominal porosity. Results from one test are summarized in Table 3.2. The data clearly indicate that the TRU isotopes exist primarily as precipitates and therefore can be removed by filtration.

Table 3.2. Filtrate and filter cake assays

Parameter	Filtrate ^a (nCi/mL)	Filter cake ^a (nCi/g)
Gross alpha	<0.05	2.64E+03
²³⁸ Pu	0.02	6.90E+02
²³⁹ Pu, ²⁴⁰ Pu	N.D.	2.40E+02
²³⁵ U	0.01	7.30E+01
²⁴¹ Am	N.D.	8.00E+00
²⁴⁴ Cm	N.D.	1.62E+03
Gross gamma ^b	1.81E+06	2.09E+07
Gross beta	4.89E+03	6.51E+05
¹³⁷ Cs	4.24E+03	1.20E+04
¹³⁴ Cs	2.10E+01	N.D.
⁶⁰ Co	8.00E+00	6.57E+03
⁹¹ Zr	N.D.	2.70E+02
¹⁵² Eu	N.D.	9.81E+03
¹⁵⁴ Eu	N.D.	6.16E+03
¹⁵⁵ Eu	N.D.	2.95E+03

^aIn this column, N.D. denotes that the isotope was not detected.

^bCounts per minute per milliliter (liquid) or counts per minute per gram (solid).

An HCTL that consists of two parallel cross-flow filters is under construction. One filter will be a bare tube unit, whereas the other tubular filter will include a zirconium oxide/silica membrane deposited on the inner surface. A granular media filter will also be tested for TRU isotope removal. Filtrate will be continuously fed through columns charged with various ion-exchange media for ¹³⁷Cs and ⁹⁰Sr decontamination studies. These studies are in direct support of the Operations Division program for LLW segregation and decontamination of the waste currently stored in the MVSTs.

Other Studies

Photographic Waste Processing. A process for recovering silver from waste photographic developer/fixer/solutions was developed at ORNL. Silver is precipitated and recovered from solution

by pH adjustment with caustic and chemical reduction with sodium hydrosulfite. The remaining solution (essentially silver free) contains an estimated 100 mg/L of undetermined organic compounds (from the original photographic solutions) and ~15 g/L of residual sodium hydrosulfite. The operation of the ORNL processing facility was halted because of a fish kill when the waste solution was discharged to a creek. This was believed to be due to a reduction in dissolved oxygen caused by the sodium hydrosulfite. It has also been determined that the waste solution is toxic to minnows, but the basis for the toxicity is not known. Alternative means of treating this waste were considered, including chemical destruction of sodium hydrosulfite with oxidizing agents and treatment in ORNL's sewage treatment plant (STP).

The disposal of the waste solution into ORNL's STP was investigated. Two small-scale treatment units were used to simulate ORNL's STP. A reduction in total organic carbon (TOC) was used as a measure of biological activity and hence the effectiveness of the test units. One unit served as a control (sanitary waste only). Photographic waste generated from a test run of the silver recovery operation was used to amend the feed to the other unit.

The addition of the photographic waste to the activated sludge test unit at up to twice the expected rate had no discernible effect on the biological activity or effluent quality as determined by measurements in TOC reduction (Table 3.3). It is not known if the organics in the photographic waste were degraded because the amount of organic carbon added to the feed stream was very low compared with that in the sanitary waste. The toxicity of the photographic waste to the biological population in the test unit at a feed rate four times that of the projected operating rate was indicated by a change in the physical appearance of the sludge (i.e., it became much lighter in color) and a significant reduction in the sludge volume.

Forty-eight-hour fat head minnow (*Pimephales promelas*) acute biotoxicity tests performed on the effluents from both the control and photographic-waste-amended (equivalent of 329 L/d to STP) reactors revealed no toxicity at 100% strength. Seven-day chronic biotoxicity tests are planned. On the basis of the results of these experiments, a permit has been requested from the state of Tennessee for this disposal method.

Coal Yard Runoff Treatment Plant Study. The wastewater streams generated at the ORNL steam

Table 3.3. Total organic carbon (TOC) reduction on a control and a photographic-waste-amended activated sludge reactor

Date (1986)	TOC (mg/L)			
	Control reactor (without photographic wastes)		Photographic-waste- amended reactor	
	Influent	Effluent	Influent	Effluent
11/4	34.0	4.1	34.0	4.8
11/5	24.3	3.0	22.2	4.0
11/6	7.1	2.6	8.9	3.7
11/7	7.8	3.1	9.4	4.0
11/11	18.0	3.5	16.0	5.3
11/12	11.0	2.9	10.0	4.8
11/14	12.0	3.0	10.0	4.7
11/17	15.0	3.1	11.0	3.4
11/18	28.0	4.9	16.0	4.2
11/21	67.0	3.9	43.0	4.1
11/26	31.0	4.0	31.0	4.2
12/1	17.0	2.9	17.0	3.1
12/4*	8.8	3.4	9.0	4.5
12/8	11.0	3.2	18.0	3.2

*On this date, the photographic waste concentration was double the initial concentration.

plant are currently treated at the 3518 treatment facility, where they are neutralized with calcium hydroxide before being released to White Oak Creek. Future plans are to treat these wastewater streams at the Coal Yard Runoff Treatment Facility (CYRTF). An R&D program was

conducted to determine the feasibility of treating such streams at the CYRTF. This program included acquiring samples of the wastewater streams to be treated at the CYRTF, characterizing the samples, performing a bench-scale evaluation of the CYRTF flowsheet, and temporarily diverting the wastewater streams from the ORNL steam plant to the CYRTF and monitoring the operation of the CYRTF on the diverted wastewaters.

The five major effluent streams to be treated at the CYRTF include coal yard runoff (CYR), hydrogen ion-exchange regeneration waste, sodium ion-exchange regeneration waste, boiler blowdown, and ash rinse waste. Samples of each of these streams were analyzed and compared with the NPDES permit² discharge limits. The results of the analyses showed that the majority of components regulated do not violate the NPDES discharge limits, even before treatment. Those that exceeded the limits included the boiler blowdown stream for copper and total suspended solids (TSS), the sodium ion-exchange regeneration waste for zinc, and the CYR for Fe, Mn, and Zn, with Cu being marginal. In addition, the composition of lead and selenium in the CYR stream violated the drinking water standards before treatment.

A flow diagram of the CYRTF is presented in Fig. 3.6. The wastewater is stored in a 760,000-L

Fig. 3.6. Flow diagram of the CYRTF.

basin until enough wastewater is collected to run the CYRTF. The wastewater from the collection basin is pumped into the pH adjustment tank, where it is mixed with lime slurry until a pH of ~10.5 is obtained. After pH adjustment, the wastewater enters a flocculator/clarifier for liquid/solids separation. The solids are dewatered using a rotary vacuum filter, and the liquid enters a recycle tank, where sulfuric acid is added to adjust the pH of the wastewater to the 6-9 range. The effluent from the recycle tank flows to a discharge basin before being discharged to White Oak Creek. The facility is designed to automatically recycle the wastewater to the recycle tank should the pH recorder or turbidity meter indicate that the wastewater is out of specification (indicated by the dashed line in Fig. 3.6).

To simulate this flow scheme at the bench-scale pH adjustment, reactor/clarifier jar tests and sludge dewatering by filtration were examined on various mixing combinations likely to occur in the CYRTF with the addition of the waste streams from the steam plant. The analyses for the six mixing events simulated are presented in Table 3.4. Before any treatment, all mixing events violated the NPDES discharge limit for zinc (mixing event 1 is included because it is only 0.01 mg/L from

the NPDES discharge limit). In addition, mixing events 5 and 6 exceeded the NPDES limits for copper and iron, while mixing events 1, 3, and 4 exceeded the limit for iron. For those components which are included in the NPDES permit without a discharge limit, the pH for all mixing events, the manganese for mixing events 3, 4, 5, and 6, and the cadmium for mixing events 5 and 6 violated either (or both) the drinking water standards or the Tennessee Effluent Limitations. It should be emphasized that these violations were prior to any treatment.

The bench-scale tests indicate that, by raising the pH to 9 and providing a 30-min settling period, all components listed above as being in violation of the discharge limits were reduced to levels less than those listed in the discharge limits. Vacuum filtration tests showed that the sludges produced could be dewatered by the rotary vacuum filter currently in use at the CYRTF.

To test the operability of the CYRTF on various mixing events, the wastewater from the steam plant was diverted to the CYR collection basin and held until enough wastewater had been collected to run the CYRTF. A total of six events was monitored during this program. During these monitoring events, samples were taken from the collection

Table 3.4. Concentration of regulated pollutants in mixing events

Parameter	Concentration (mg/L) in mixing event					
	1 ^a	2 ^b	3 ^c	4 ^d	5 ^e	6 ^f
pH	2.0	1.9	2.1	2.0	2.5	2.5
TSS	8	10	12	34	27	
Cr	<0.080	<0.080	<0.080	<0.080	<0.080	<0.080
Cu	0.66	0.45	0.69	0.39	1.1	1.1
Fe	1.1	0.30	>53	59	110	210
Zn	0.99	1.4	1.4	1.3	1.8	1.8
As	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20
Cd	<0.010	<0.010	<0.010	<0.010	0.016	0.016
Pb	<0.40	<0.40	<0.40	<0.40	<0.40	<0.40
Mn	0.025	0.026	0.81	0.47	1.1	1.7
Ni	<0.12	<0.12	0.24	0.16	0.43	0.49
Se	<0.40	<0.40	<0.40	<0.40	<0.40	<0.40
Ag	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10

^a48.9% ion-exchange acid regeneration (IXA), 13.3% ion-exchange sodium regeneration (IXS), 28.8% boiler blowdown (BB), 9.0% ash rinse (AR).

^b53.7% IXA, 14.6% IXS, 31.7% BB.

^c2.8% IXA, 7.3% IXS, 15.9% BB, 50.0% CYR.

^d40.2% IXA, 11.0% IXS, 23.8% BB, 25% CYR.

^e100% CYR (obtained July 9, 1986).

^f100% CYR (obtained July 14, 1986).

basin, the recycle tank, and the effluent weir draining into White Oak Creek. The resulting data showed that only iron violated the NPDES permit. This was attributed to operational problems occurring with the flocculator/clarifier throughout the monitoring period. Solid samples from the rotary vacuum filter contained 65 to 75% moisture. Results from the Extraction Procedure (EP) Toxicity Test¹¹ indicated that all of the elements analyzed for in this test were below levels considered EP-Toxic.

Laundry Waste Treatment. A study is under way to determine ways of treating waste solutions generated by laundering radioactively contaminated clothing at ORNL. A literature survey indicated that several potential processing alternatives (e.g., RO chemical precipitation, ion exchange, evaporation, and centrifugation) are applicable, but successful use has been site specific and is highly dependent on the type of detergent used. Laboratory-scale tests are being performed to determine the feasibility of treating laundry wastes at the PWTP or by RO.

Four types of detergents were tested to determine the effects on PWTP operation: (1) Turco 4324,^{*} the phosphated detergent presently used at the ORNL laundry; (2) Turco 4324-NP,^{*} a non-phosphated detergent developed for nuclear applications; (3) Mark detergent, a heavy-duty non-phosphated industrial laundry detergent manufactured by U. N. X. Chemicals, Inc.; and (4) Diversey Wyandotte F-901,[†] a liquid surfactant that does not contain detergents. Results of laboratory-scale tests indicated that both Turco detergents will inhibit the water-softening step at the PWTP. The data suggest that the Mark and Diversey Wyandotte detergents will not significantly affect the softening process or sorbents being used or considered for use at the PWTP, that is, chabazite zeolites, IRC-84 resin, and HCR-S resin.

In RO tests, effluent samples from the ORNL laundry containing the Mark and Diversey Wyandotte detergents were passed through a 50-mm, spiral-wound, cellulose acetate membrane. The radionuclides and metals were rejected by the membrane, and no gross fouling of the membrane was detected during 9 h of operation.

Tests were performed at the ORNL laundry to determine whether the Mark and Diversey Wyandotte detergents will effectively clean the clothing. Although both detergents removed alpha and beta-gamma contamination relatively well, the

Diversey Wyandotte detergent did not adequately clean the clothing. Additional testing of the Mark detergent has been suggested.

3.2 BIOPROCESSING SUPPORT TO THE FEED MATERIALS PRODUCTION CENTER

The Feed Materials Production Center (FMPC) at Fernald, Ohio, is constructing a fluidized-bed bionitrification plant based on pilot work conducted at ORNL in the late 1970s and early 1980s. This plant is designed to treat approximately 600 to 800 L/min of wastewater having a nitrate concentration as high as 10 g/L. The effluent is to contain less than 0.1 g/L of nitrate. Four fluidized-bed bioreactors in series, each 1.3 m in diameter and 12 m tall, will be used.

Since this new facility is an extrapolation of the ORNL work to significantly larger scale equipment and to actual rather than synthetic wastewater, design verification studies have been performed at ORNL to reduce uncertainties in the scaleup.

The 12-m-tall bioreactors at the FMPC were simulated using two different bioreactor systems at ORNL. One system contained two 10-cm-diam, 6-m-tall glass pipe bioreactors in series. The other consisted of a single 10-cm-diam, 12-m-tall bioreactor constructed from polyvinyl chloride pipe. Both systems had sample ports located approximately every 1 to 1.3 m along the length of the bioreactors for axial sampling and could be operated with varying recycle rates or once-through treatment of wastewater. Off-gas was vented through wet-test meters connected to recorders to produce continuous records. Effluent wastewater and entrained bioparticles were passed through a vibrating screen system to shear off excess biofilm and to recover the bioparticles for recycle to the column. A schematic of the two-column system is shown in Fig. 3.7.

Various combinations of synthetic wastewater and actual wastewater shipped to ORNL from the FMPC were treated over a 9-month period. The major findings during these studies are summarized in the following paragraphs.

11. Code of Federal Regulations, Vol. 40, No. 141.

^{*}Manufactured by Turco Products, Inc.

[†]Manufactured by Diversey Wyandotte Corp.

Fig. 3.7. Schematic diagram of the two-bioreactors-in-series pilot facility.

- Wastewater from the FMPC is biotreatable at sufficient rates to achieve the design performance of the bionitrification plant. This design rate is 32 kg of nitrogen per cubic meter of bed volume per day. The off-gas rate from the bioreactors is a reliable indicator of the real-time performance of the system and correlates well with the nitrate destruction rate based on nitrate assays. Both methanol and ethanol worked well as carbon sources. (Theoretically, the reaction stoichiometry will be different for methanol than for ethanol, which should alter the quantity of hydroxyl produced and thus affect the pH shift. However, this effect was not carefully examined in these studies.)
- Metered addition of acid within the bioreactor at selected locations appears to be an effective means of mitigating the pH rise and thus maintaining high reaction rates. Sulfuric, acetic, and phosphoric acids were used in these studies at ORNL with equal performance. The amount of acid required, and perhaps the need for acid addition at all, depends on the natural buffering capacity of the wastewater and on the particular carbon source, as noted above.
- Calcium in the wastewater leads to precipitation of calcium carbonate on the bioparticles and elsewhere in the bioreactor system. If this precipitation is excessive, it shuts down the biological action by covering over the bioparticles. This problem was experienced severely with FMPC wastewater containing 300 to 400 mg/L of calcium. Biological destruction ceased within 1 week of exposure to this wastewater, and the system had to be restarted. However, softened wastewater was treated successfully on a long-term basis. Evidence from these studies suggests that a calcium level of ~50 mg/L will probably be acceptable; concentrations significantly higher than 50 mg/L may lead to problems.
- Daily removal/replacement of approximately 2 to 5% of the bed volume with fresh coal was shown to be an effective, and perhaps necessary, action to maintain high reaction rates. This action supplements the role of the vibrating screens in maintaining thin biofilms on the coal. Without removal and replenishment, the biofilms tend to overgrow and create large bioparticles that are ineffective catalysts, analogous to internal diffusional limitations in classical porous

catalysts. Addition of fresh coal also serves to ensure that bioparticles are carried over into the vibrating screens for stripping of the excess biofilm.

- The temperature rise created by the exothermic reaction is of the order of 0.5°C per meter of bioreactor height at the design reaction rate for the FMPC plant. Experimental results and theoretical predictions are shown in Table 3.5 for several periods in which the experimental biodenitrification rates were stable over several days. The free energy of reaction has been reported to be -138.6 kcal per mole of methanol, which is the basis for the theoretical calculations. For the total bioreactor height of 48 m at the FMPC, the total temperature rise (under adiabatic conditions) would be about 20°C . Temperature control of some sort may be needed to maintain an acceptable temperature for the biological activity. However, the temperature dependence of the biodenitrification rate is not well characterized.

Table 3.5. Comparison of the theoretical and measured temperature rise in the 12-m pilot bioreactor at various denitrification rates

Denitrification rate (kg N/d·m ³)	Theoretical temperature rise ($^{\circ}\text{C}$)	Experimental temperature rise $\pm$ standard deviation ($^{\circ}\text{C}$)
8	1.5	0.93 ± 0.50
20	3.7	3.59 ± 0.52
30	5.5	4.46 ± 1.11

Source: J. F. Walker, Jr., M. V. Helfrich, and T. L. Donaldson, *The Biodenitrification Development Program*, ORNL/TM-10239, February 1987, Table 6, p. 43.

Biodenitrification is expected to be an effective technology for treatment of the nitrate-containing wastewaters at the FMPC. Further testing with the full-scale bioreactors at the FMPC will be used to determine the particular operating conditions necessary to achieve and maintain the desired biodenitrification rate. The only significant accommodation for biotreatment of the wastewater appears to be restriction of the calcium concentration to ~ 50 mg/L. Several strategies are being considered by FMPC personnel to meet this requirement.

The biodenitrification plant is scheduled for startup during the spring or summer of 1987. Chem Tech personnel have assisted in the design of the startup program and will assist with on-site process engineering expertise during the initial demonstration campaign.

3.3 BIOOXIDATION OF COAL GASIFICATION WASTEWATERS

This experimental program has been pursued for several years with funding from the Morgantown Energy Technology Center (METC). Recent emphasis has been on characterization of the biooxidation of individual major organic pollutants in wastewaters obtained from METC and on enhancement of bioactivity by supplemental inoculation with specific microbial strains. An assessment of wastewater treatment technologies for coal conversion wastewaters was also initiated.

The bench-scale bioreactor (Fig. 3.8) used for these studies consisted of a tapered section with a 1.27-cm ID at the bottom and a 2.54-cm ID at the top. The tapered section was about 25 cm long and contained about 100 mL total volume. A 1-L (300-mL working volume) reservoir was used for mixing of fresh feed and oxygen sparging.

The wastewater used in these experiments was obtained from METC in 1985 (producer 102,

ORNL DWG 86-81

Fig. 3.8. Schematic of the experimental bioreactor. Source: T. L. Donaldson, D. D. Lee, and S. P. N. Singh, *Treatment of Coal Gasification Wastewaters: Final Report*, ORNL/TM-10241, February 1987, Fig. 1, p. 3.

Blacksville Coal, October 3, 1983). Most of the sulfides and ammonia were stripped from the water using steam and nitrogen. The solid support for the microbial films was 30- to 60-mesh anthracite coal, washed to remove the fines. The microbial culture was obtained from previous pilot-scale fluidized-bed bioreactors used for development of this process.

Phenols were measured using the 4-aminoantipyrine (4-AAP) method and capillary gas chromatography. Extraction of organics for chromatography was carried out using methylene chloride. Pure compounds were used to determine the retention times for the following species of interest: phenol; *o*-, *m*-, and *p*-cresols; 2,3-, 2,4-, 2,6-, 3,4-, and 3,5-xylenols; 2-, 3-, and 4-ethylphenol; resorcinol; catechol; naphthalene; phenanthrene; and anthracene.

The bioreactor was started on dilute phenol and mineral salts media, and METC water was added gradually over a period of several weeks until the feed to the fluidized bed contained ~1% METC water.

Following biooxidation in the fluidized-bed bioreactor system, the 4-AAP-phenols content was typically reduced by 40 to 50%, depending on the residence time. Concentrations and degradation rates for various specific compounds are shown in Table 3.6 from a typical sample taken ~3 months after startup of the experimental program. These concentrations were measured by gas chromatography at the inlet and outlet of the fluidized bed. The data show that monohydric phenol and *m*-, *p*-cresol are degraded most rapidly, while *o*-cresol, 2,3-xyleneol, and 3,4-xyleneol are degraded an order

of magnitude more slowly. Other organics are degraded even more slowly. These relative rates are typical for the data obtained in this system.

It was generally observed that monohydric phenol was degraded first, with relatively little simultaneous degradation of other compounds. However, when the liquid residence time was sufficiently long to degrade most of the phenol, the degradation of the other compounds increased. This behavior is typical of microbial degradation of mixed substrates. In this case, phenol is the simplest substrate and the most favorable energetically for the microorganisms. The catabolic pathways for the more substituted aromatic rings are more complex, more energetically demanding, and kinetically slower.

Gene probe assays were done in collaboration with Gary S. Sayler, of The University of Tennessee. The presence of the genotype for naphthalene degradation was assayed using Sayler's probe for the NAH7 plasmid. After some initial characterization work on the microbial population in the bioreactor, an experiment was conducted to determine if a new microbial culture could be established in the existing bioreactor system. A culture containing the NAH7 plasmid (obtained from Sayler) was added to the bioreactor system, and the liquid residence time was increased for 1 d to reduce the washout of free cells. Then the residence time was returned to normal conditions. Samples of biofilm were obtained before the inoculation and at 1, 3, and 5 d after inoculation for gene probe analysis to determine the level of the NAH7 plasmid.

Results of the gene probe assays showed that the

Table 3.6. Organic components measured by methylene chloride extraction and gas chromatography (typical analysis)

Component	Inlet to fluidized bed (mg/L)	Outlet from fluidized bed (mg/L)	Degradation rate [mg (L bed volume) ⁻¹ min ⁻¹]
Phenol	72	66	10
<i>o</i> -Cresol	59	58	1.7
<i>m,p</i> -Cresol	114	106	14
3,5-Xylenol	4	4	0
2,3-Xylenol	24	22	3.4
Naphthalene	7.9	7.6	0.5
3,4-Xylenol	49	48	1.7
Catechol	1.2	1.2	0

Source: T. L. Donaldson, D. D. Lee, and S. P. N. Singh, *Treatment of Coal Gasification Wastewaters: Final Report*, ORNL/TM-10241, February 1987, Table 2, p. 9.

initial mixed population in the bioreactor already contained the NAH7 plasmid. This is not particularly surprising since it is known that this and similar plasmids are relatively common in natural microbial populations. After inoculation, the assay showed no significant change in the quantity of this genotype in the biofilm. However, the naphthalene degradation rate increased by an order of magnitude, and this increased rate was maintained for 2 weeks until the bioreactor system was shut down. Such behavior certainly suggests that the introduced microorganisms were able to colonize the existing biofilms to some extent. In addition, the phenol degradation rate appeared to be somewhat improved after inoculation; this would be expected since the NAH7 plasmid also contributes to the metabolic pathway for phenol degradation. These results are encouraging and indicate that more studies should be done.

A comprehensive assessment of the technology used to treat coal gasification wastewater was initiated in FY 1986. The published literature was reviewed and a bibliography prepared. Discussions were held with several other research groups working on treatment of coal gasification wastewaters, and a visit was made to the Great Plains coal gasification plant at Beulah, North Dakota, and to the University of North Dakota Energy Research Center at Grand Forks. No additional funding was provided in FY 1987 to complete the assessment; thus the project has been terminated.

3.4 WATER QUALITY PROGRAMS

EPA Water Quality Program: Evaluation of Bioassays Used to Estimate Health Hazards of Wastewater Effluents

This research program was developed through an interagency agreement with the U.S. Environmental Protection Agency (EPA) in collaboration with the ORNL Analytical Chemistry, Chem Tech, Environmental Sciences, and Health and Safety Research divisions. The major objective of this interdivisional program is to evaluate bioassay methods for predictive capabilities relative to human health hazards associated with the discharges of wastewater treatment plants or industrial effluents. This program requires a detailed and thorough examination of selected effluents using a battery of in vitro bioassays in combination with chemical characterization. Bioassays include

the Ames mutagenicity test with *Salmonella typhimurium* strains and several Chinese hamster ovary (CHO) cell tests. Sample collection, preservation, storage, and concentration techniques are being examined to determine optimum methods to concentrate and store samples while minimizing sample degradation and alteration. Bioassay information will be coupled with chemical characterization in order to develop a more complete understanding of the chemical nature of possible toxicants and the potential use of bioassays for estimating human health risk associated with wastewater discharges. The role of the Chem Tech Division in this multidivision effort was to collect, process, and preserve the wastewater samples.

Several wastewater treatment plants were evaluated for possible effluent sampling and study in this research program. The selection criteria included the known or potential toxicity, mutagenicity, or carcinogenicity of treatment plant effluent; well-characterized and capable plant treatment and management; and a cooperative attitude relative to sampling assistance. An additional, but not essential, criterion was an ongoing chemical and toxicological characterization effort at the plant which would permit integration of our research findings into a larger, more inclusive characterization effort. Two treatment plants were selected for further study: (1) Sauget Wastewater Treatment Plant, Sauget, Illinois, and (2) Moccasin Bend Wastewater Treatment Plant, Chattanooga, Tennessee. The Sauget plant is a physical-chemical treatment plant, and the Moccasin Bend plant is a primary and secondary wastewater treatment plant using a biologically activated sludge process.

The wastewater treatment plant at Sauget, Illinois, was selected for intensive studies in this research program. This plant uses physical-chemical treatment to process daily 9 million gallons (34 million liters) of predominately industrial waste from the East St. Louis area, which includes chemical plants, electroplating plants, and oil refineries. The treatment plant uses the following unit operations sequentially: oil skimmer; sand and grit removal after partial pH adjustment by the addition of lime; further adjustment to pH 8.3 by the addition of lime; flocculation of precipitated heavy metals assisted by the addition of a long-chain anionic polymer; coagulation; settling; sludge removal; and discharge of the treatment

plant effluent to the Mississippi River. Approximately 20,000 gal (76,000 L) of oil per year is skimmed from the influent and shipped to an off-site hazardous waste incinerator. The sludge is mixed with lime, dehydrated, and shipped to a hazardous waste landfill. Sludge disposal requires a hazardous waste permit from the state of Illinois.

On April 12, 1985, preliminary contacts were made at the Sauget site. Initial samples (23 L of effluent) were collected, quick-frozen on dry ice, and shipped by air to ORNL. Sampling protocol, equipment requirements, and methods for collecting large sample volumes (up to 1,000 gal, or 3,800 L) were discussed with plant personnel.

On May 10, 1985, twelve 38-L influent samples and twelve 3.8-L "archive" influent samples were collected in polyethylene containers, and 12 analytical samples of influent were collected in specially cleaned glass containers. The archive samples were quick-frozen on dry ice. The influent sample site was located immediately upstream from the lime neutralization treatment. The samples were collected by using an immersible pump operating at the rate of 2 gal/min (7.6 L/min). The timing of effluent sample collection was calculated to be representative of the wastewater at the time of influent sample collection based upon plant residence time. The following effluent samples were collected immediately downstream from the settling basins: twelve 55-gal (208-L) samples in stainless steel drums, twelve 1-gal (3.6-L) archive samples in polyethylene containers, and 15 analytical samples in specially cleaned glass containers. The drum samples were collected by using the wastewater treatment plant's recirculating pump. The archive and analytical samples were obtained as grab samples. The archive samples were quick-frozen on dry ice.

All Sauget samples (about 750 gal, or 2,800 L) were transported by refrigerated truck to ORNL. The 11 drum samples of effluent were blended. Aliquots of the blended sample were subjected to different treatments: rough filtered (10 μ m), stored at 2°C or -10°C, acidified to pH 2, ultrafiltered (0.22 μ m), or partially lyophilized. Samples of these aliquots were subjected to a battery of bioassays. The analytical samples were examined by using several different organic and inorganic analytical procedures. The bioassay and analytical results are presented elsewhere.¹² In general, the Sauget effluent was mutagenic in *S. typhimurium* strains TA98 and TA100 with and without S9

activation. At the volumes tested (up to 3 mL), there was no mutation produced in the CHO assay and survival was reduced by only 30%. A large number of organic compounds were detected in the effluent, including benzene, toluene, ethylbenzene, chlorobenzene, dichloroethanes, trichloroethanes, tetrachloroethane, dichloropropane, dichloropropenes, methylene chloride, carbon tetrachloride, chloroform, bromoform, and mixed bromo-chloro-haloforms. Concentrations in the effluent were determined for the following compounds: aniline, 105 μ g/L; 4-chloroaniline, 29 μ g/L; 2-nitroaniline, 229 μ g/L; 3- or 4-nitroaniline, 34 μ g/L; diethyl phthalate, 1 μ g/L; dibutyl phthalate, 6 μ g/L; and butylbenzyl phthalate, 10 μ g/L. Analytical results and bioassay results were determined by the Analytical Chemistry and Health and Safety Research divisions, respectively.

The Moccasin Bend Wastewater Treatment Plant was selected for additional studies to determine the applicability of methodologies developed using the Sauget wastewater samples. The plant serves the city of Chattanooga and several outlying communities in Tennessee and Georgia. This secondary wastewater treatment plant treats up to 65 million gallons (246,000 m³) of wastewater per day using a cryogenic pure-oxygen-activated sludge process. About half the dry-weather flow comes from 120 major industrial sources that contribute about 65% of the influent biochemical oxygen demand and about 75% of the chemical oxygen demand. Seventy-three of the EPA's 129 priority pollutants have been detected in the plant influent.¹³ In the wastewater treatment plant, the following steps are used: conventional grit removal, primary settling, secondary activated sludge treatment with oxygen, secondary settling, chlorination, and discharge of the chlorinated effluent to the Tennessee River. The sludge is dehydrated and disposed of in a landfill.

On November 13, 1986, preliminary contacts were made at the Moccasin Bend plant. Sampling protocol and scheduling and cooperative efforts were discussed. On November 21 and 22, 1986, influent and effluent samples were collected. Ten

12. R. L. Schenley, L. W. Condie, M. V. Buchanan, R. L. Jolley, L. C. Waters, B. H. Owens, and P. J. Walsh, "Complex Mixtures: No Simple Solutions," submitted to *J. Toxicol. Environ. Health*.

13. G. E. Kurz and R. J. Henderson, "A Rapid Industrial Waste Screening Method," presented at the 39th Purdue Industrial Waste Conference, Lafayette, Ind., May 9, 1984.

1-gal (3.8-L) grab samples and two 13-gal (49-L) 24-h composite samples of the influent were collected and stored in polyethylene containers. The 1-gal archive samples were quick-frozen in the plant freezer. Analytical samples were also collected on XAD resin and on columns containing Blue Cotton (manufactured by Funakoshi Pharmaceutical Co., Ltd., Tokyo, Japan). Timing of the effluent samples was calculated to correspond to the plant residence time of the influent when sampled. Effluent samples immediately upstream of the chlorinator were collected as follows: ten 1-gal (3.8-L) archive samples in polyethylene containers, two 13-gal (49-L) 24-h composite samples in polyethylene containers, and 2-L samples adsorbed on XAD and Blue Cotton. In addition, a 1-gal sample and several small analytical samples of the effluent were collected after chlorination. These samples are being evaluated using selected bioassays.

3.5 IN SITU BIOTREATMENT OF HAZARDOUS WASTES

A new cross-disciplinary, multidivisional program has been initiated with support from the Laboratory Director's R&D Fund. The work is focused on fundamental questions in microbial ecology in the context of biotreatment of hazardous wastes. Critical issues being addressed include the survival of microorganisms introduced to hazardous waste environments, the retention of desirable functional properties, gene stability, movement of the microorganisms in natural media, and the potential for transfer of genetic material between species. Improved understanding of these issues is critical both to the development of workable technologies and for analysis of the risks associated with introduced microorganisms. This program has already attracted additional funding from the U.S. Air Force for decontamination of groundwater, and serious discussions for additional new projects are under way with other potential

sponsors. Personnel participating in this program are from the Chem Tech and Environmental Sciences divisions of ORNL and The University of Tennessee.

Biodegradation of trichloroethylene (TCE) to CO_2 and water-soluble products by methylotrophic microorganisms isolated from wells at the Y-12 Plant has been demonstrated using ^{14}C -TCE. The growth kinetics of an isolate from this culture depend on the concentrations of methane, copper, and TCE. Polyclonal antiserum was developed against this isolate and was shown to react positively with a variety of other methanotrophs and with free-living amoebae isolated from Y-12 wells and other sites. This suggests previous interactions of the amoebae with methylotrophs and has implications for population dynamics in natural, open environments containing amoebae.

A mathematical model of microbial growth kinetics was developed and integrated with an existing groundwater transport model. The growth kinetics of the TCE-degrading isolate were used to calibrate the biological component of the integrated model.

A model denitrifying biofilm has been established with *Pseudomonas chloroaphis* ATCC 9446 and *Pseudomonas* sp. ATCC 13867 for population dynamics studies in a laboratory bioreactor. These organisms have different fatty acid profiles, which offer an attractive, rapid technique to differentiate between the two species in mixed culture. Development of specific techniques to monitor the biofilm populations based on the fatty acid assays has been initiated.

Extensive interactions with personnel at The University of Tennessee led to preparation of a proposal to the National Science Foundation's Engineering Research Centers Program to establish the Center for Environmental Biotechnology at ORNL and The University of Tennessee. This concept has been the focal point for many productive discussions with potential sponsors, and development of the Center is being pursued.

4. BASIC SCIENCE AND TECHNOLOGY

4.1 CHEMICAL SCIENCES

Chem Tech's research in the Basic Energy Sciences and Basic Chemical Sciences programs includes a series of fundamental studies, mostly relating to various aspects of chemical separations. These studies are direct outgrowths of the Division's historical expertise in separations of uranium, fission products, and other radioisotopes in nuclear fuels and feed materials. In recent years our research programs have diversified and are generally of a more basic nature, focusing more often on specific aspects of separations processes than on a general separations method.

For example, one current study in the Chemical Sciences Program is concerned with the kinetics of selected reactions that are catalyzed with enzymes or other biologically based materials. This is a growing research effort which has afforded new insight into biophotochemical systems that are of particular interest for their potential energy applications.

Chemistry of Actinides and Fission Products

This study's primary purpose is to explore fundamental chemistry of the actinide elements and their fission products. Specific areas of interest include: (1) photochemical studies of U, Np, and Pu in aqueous solution; (2) characterization studies of the structures and properties of pure and mixed actinide polymers; (3) spectroscopic and photochemical investigations of actinides in low-temperature matrices, with particular emphasis on isotope effects; (4) kinetic and thermodynamic studies of complexation and redox reactions in aqueous and nonaqueous solutions; (5) properties of certain complexes with macrocyclic ligands; and (6) studies of fundamental properties of aqueous and nonaqueous solvent systems in support of these primary fields of investigation.

Studies of Actinide Polymers. Small-angle X-ray scattering (SAXS) experiments have been initiated to characterize hydrous polymers suspended in solution. To date, only optical spectra of the polymers in organic media have been reported in the literature by other workers, and the ambiguity of the spectral assignments to the polymer form has resulted in considerable uncertainty regarding the extractability of these polymers. The most reliable resolution of this problem would be identification of the polymer using an experimental approach that addresses the size of the particles. Of several suitable techniques, small-angle scattering (both X-ray and neutron) offers a direct means for studying hydrous inorganic polymers in both aqueous and organic media over a size range that is characteristic of the initial polymerization stages. Because of its availability and applicability, we have selected SAXS and have used it to characterize the size distribution of polymers as a function of pH and aging conditions.

Thorium(IV) polymer suspensions were examined first, because they are typical of the actinide polymers and may be studied without alpha containment. We have found that freshly prepared Th(IV) polymer, which is formed from a 0.1 M Th(NO₃)₄-0.1 M HNO₃ solution by titration with NaOH to pH 4.0, produces a colloidal suspension with polymeric aggregates having diameters of 2.6 to 5.5 nm (Fig. 4.1). Aging by reflux for a period of 1 to 3 h causes further aggregation, with colloidal particles of up to 43-nm diam in aqueous solution (Fig. 4.2). When these suspensions were extracted with 0.5 M dibutyl phosphate dissolved in *n*-dodecane, colloidal particles of up to 12.6-nm diam were found in the organic phase, while larger colloidal particles precipitated. None of the other organics that have been reported to extract hydrous polymers, for example, trioctylphosphine oxide and dihexyl[(diethylcarbamoyl)methyl]phosphonate, were found to extract the Th(IV) polymer.

Fig. 4.1. Small-angle X-ray scattering isointensity contour map showing the scattering from smaller (2.6- to 5.5- μm -diam) polymer particles as seen by an array detector at a distance of 2.126 m from the sample, which was irradiated with K_α X-rays from a copper source.

These results have demonstrated that hydrous polymers can be extracted into, or suspended in, organic phases under selected conditions. However, the capacity of the organic extractant for the polymer is not as great as is that of the aqueous phase. Nevertheless, the potential extractability of these polymers must be considered in practical separations processes, and the results of these experiments should be useful in explaining the nature and extent of this polymer behavior.

Actinide Photochemistry. During FY 1986, spectroscopic and photochemical studies of matrix-isolated actinide species were begun. The purpose of this work is to provide detailed information about the fundamental processes which occur during actinide photo-redox reactions. The reaction chosen for initial study is the photochemical reduction of uranyl ion (UO_2^{2+}) by methanol and visible

light. A significant part of our effort was devoted to developing the experimental hardware and techniques necessary for producing pure dilute samples of highly reactive gases and to establishing the operating limits of our instrumentation. Our infrared spectrometer is able to record spectra of matrix-isolated species at 1:1000 dilution, with a maximum resolution of $\sim 1 \text{ cm}^{-1}$. While this is adequate for the current task of identifying reactant and product species, greater precision will be required if actinide isotope splittings are to be measured.

The original study included the simultaneous production of the two matrix-isolated reactants, $\text{UO}_2(\text{OCH}_3)_2$ and CH_3OH , by the hydrolysis of uranium hexamethoxide, $\text{U}(\text{OCH}_3)_6$. This is a slightly volatile, easily hydrolyzed solid which, unlike UF_6 , does not attack glass or metal. After

Fig. 4.2. Small-angle X-ray scattering isointensity contour map showing the scattering from large (14.4-nm-diam) polymer particles as seen by an array detector at a distance of 2.126 m from the sample, which was irradiated with K_α X-rays from a copper source.

synthesizing the compound and obtaining the infrared spectrum of a flash-evaporated thin film, we conducted a series of experiments with argon and krypton matrices and varying deposition conditions. The spectra obtained were consistent with those literature values reported for the solid compound.

In the thin film and matrix spectra, the presence of a single U-O stretching vibration at 465 cm^{-1} and the absence of the Raman active bands at 496 cm^{-1} and 401 cm^{-1} indicated that, as expected, the pseudooctahedral symmetry of the molecule was preserved. However, there were bands attributable to organic decomposition products present to varying degrees in all the spectra. Two of these have strong infrared absorptions in the 900 to 1000 cm^{-1} region, which must be free

of interfering absorptions if we are to detect isolated uranyl species. After several unsatisfactory attempts to produce matrices free of these absorptions, we concluded that the thermal stability and volatility of $\text{U}(\text{OCH}_3)_6$ were insufficient for our studies and that these properties had been somewhat exaggerated in the literature. We then shifted our attention to UF_6 .

Although it is well known that UF_6 and H_2O react in the gas phase, producing HF and UO_2F_2 , the literature gives no indication that the reaction kinetics have been studied under the dilute conditions of our experiment. We have observed that no appreciable hydrolysis occurs when the reactants, dilute in argon or krypton, are codeposited or are mixed immediately prior to deposition. At present, we are preparing to monitor the gas-phase reaction

spectroscopically to determine the appropriate intervals for mixing and reaction.

Properties of Aqueous and Nonaqueous Systems. Lanthanide and Actinide Coordination Chemistry. These studies are a continuation of collaborative work with Dr. G. R. Choppin, of Florida State University, with funding support for one postdoctoral student and one undergraduate. The studies emphasize actinide and lanthanide redox and coordination chemistry in concentrated electrolytes and low-melting fused salts. Currently the aluminum chloride-*n*-butyl pyridinium chloride and phosphoryl trichloride-tin(IV) chloride systems are under investigation. The first year of the project was spent collecting literature on these two systems and building the glove boxes and other equipment needed. A series of shakedown experiments on thermodynamics of the fluoride complexation of U(VI), Th(IV), and Nd(III) was performed to calibrate the instruments and train the students. New methods for the rapid, economical preparation of ultrapure aluminum chloride and *n*-butyl pyridinium chloride were developed.

Solvent Extraction of Lanthanides and Actinides. The goals of this task are the development and characterization of synergistic solvent extraction systems for separating the trivalent lanthanides and actinides. The work is being carried out in collaboration with Dr. D. D. Ensor, of the Tennessee Technological University. Initial studies of the mechanism for extraction of U(VI) using thenoyltrifluoroacetone (TTA) in combination with 4,13-didecyl-1,7,10,16-tetraoxa-4,13-diazacyclooctadecane (22DD) and 4-decyl-1,7,10,16-tetraoxa-4,13-diazacyclooctadecane (22D) were completed, and a report is being prepared. The most interesting result is that nitrogen atoms in the macrocycle seem to be largely responsible for the synergistic effect; analogous crown complexes are ineffective. Studies still in progress with other actinide and lanthanide ions suggest that the synergistic effect varies directly with the ionic radius. As details of this size-selective synergism are better understood, improved separation schemes for these elements will be developed.

Separation Chemistry of Transplutonium Elements in Alkaline Media. This task, exploring the separation chemistry of the transplutonium elements in alkaline media, is being done in collaboration with Dr. J. R. Peterson, of the University of Tennessee. Initial studies have focused on the chemistry of Bk(IV) and a lanthanide analogue,

Ce(IV). Evidence in the literature that amines (e.g., Aliquat 336, decylamine) can be used to extract berkelium from highly alkaline carbonate solutions was shown to be unreliable. However, it was found that trivalent lanthanide elements could be extracted from alkaline solution with alkyl-substituted catechols. The readily available catechols were too water soluble to act as good solvent extraction reagents, so a search is under way for a less soluble derivative.

Multiphase Studies in External Fields

Several fundamental studies concerned with mass transport in multiphase separations are included in this program. The overall objective is to gain a better understanding of the principles of chemical separations and thereby discover new methods for attacking the intricate problems posed by multiphase separations. The study of two-phase, liquid-liquid systems remains the current focus of activity in this area of the program. Several studies are investigating the hydrodynamics of a liquid-liquid system interacting with an electric field and how these interactions affect transport processes.

Droplet Formation in Electric Fields. Formation of the dispersed phase in a liquid-liquid system is quite often the most important contributing factor to the performance of the mass transfer operation. Previously, we reported experiments in which droplet size control was effected through the use of applied direct-current electric fields. Aqueous droplets are formed at an orifice in a square column filled with an organic nonconducting liquid. An electric field is produced between the orifice plate and a flat screen made of conducting wire. Drops formed by a slow flow rate through the orifice are subject to the following force balance between the surface force (F_s) and the sum of the gravitational force (F_g) and the electrostatic force (F_E):

$$F_s = F_g + F_E .$$

The presence of an electrostatic force allows the balance to be satisfied with smaller drops, which results in more surface area for mass transfer.

A theory based on treating the formed droplets as spheres in a uniform electric field grossly underestimated the reduction in droplet size due to the field. Hence, a new model has been developed

which calculates the force field around the drop between a charged disk (the face of the orifice plate) and a grounded infinite plane (the grounded screen electrode).

The potential (V), current flow (S), and conductivity (k) are related by

$$V = \frac{S}{2\pi k r_1} \int_0^\infty \frac{\sinh(\lambda z)}{\cosh(\lambda a)} \sin(\lambda r_1) J_0(\lambda r) \frac{d\lambda}{\lambda}.$$

The field intensity may be obtained as

$$\frac{\pi dV}{\pi dz} = E_z = \frac{S}{2\pi k r_1} \times \int_0^\infty \frac{\sinh(\lambda z)}{\cosh(\lambda a)} \sin(\lambda r_1) J_0(\lambda r) d\lambda,$$

where J_0 is a Bessel function and l is a dummy variable. The electrostatic force is given by

$$F_E = \frac{1}{2} \int \epsilon E_z^2 dA,$$

where A = area of the disk, m^2 .

A computer program was written to calculate the electrostatic forces on drops for any specified sizes and eccentricities. Results of these calculations are shown, along with experimental results for the water-cyclohexane system with an orifice diameter of 0.0317 cm, in Fig. 4.3. Investigation is proceeding for additional orifice sizes and chemical systems.

Surface Tension in Conducting Systems. To analyze the effects of electric fields on liquid-liquid systems, values for the controlling physical properties of the two fluids involved must be known. In the systems studied by Chem Tech researchers, interfacial tension is one of the properties which must be accurately evaluated under experimental conditions. Video analysis equipment makes it possible to accurately measure this property, using the pendant drop method. An algorithm for determining surface and interfacial tension from videotaped profiles of pendant drops has been developed. The program numerically integrates (fourth-order Runge-Kutta method) the Bashforth-Adams equa-

Fig. 4.3. Calculated and experimental values for drop volume in the water-cyclohexane system.

tions and uses a nonlinear parameter estimation technique to minimize the differences between the vertical positions of experimental and theoretical curves. The program was originally written in Microsoft BASIC for operation on the IBM PC/AT computer; it was later translated into FORTRAN 77 and ported to a mainframe computer (VAX 8600) to achieve a 30-fold increase in speed. The program requires ~6 min to converge if 120 data points are used; however, only 30 to 50 data points are needed to achieve a precision of within <1%, so convergence for normal usage occurs in <2 min. Furthermore, a precision of within <1% can be achieved using high-speed video cameras and high-resolution video digitization boards. Figure 4.4 shows results for the determination of the interfacial tension of a system containing an aqueous salt and 2-ethyl-1-hexanol ($\gamma = 14.1 \pm 0.08 \text{ mN/m}$) where the aqueous phase contains $100.1 \text{ kg/m}^3 \text{ NaCl}$. A Sony SMC-70 graphics computer with a 320×200 resolution was used to digitize the drop image.

Mass Transfer in Droplets. Descriptions provided by qualitative and quantitative experimental flow-visualization techniques have enabled us to develop

Fig. 4.4. Data for interfacial tension in a liquid-liquid system containing aqueous salt and 2-ethyl-1-hexanol. These data points represent a digitized profile of the pendant drop.

a detailed model for flow patterns in a droplet-continuum system typical of liquid-liquid solvent extraction. The streamlines for flow in and around circulating droplets which are forced into regular oscillation by a pulsing electric field can be modeled as the vectorial sum of low-Reynolds-number internal circulation (Hadamard flow) and an oscillatory streaming caused by the prolate-to-oblate ellipsoidal shape changes of the droplet (Dirichlet flow). The convection-diffusion equations have been derived and numerically solved for mass transfer from both circulating droplets (Hadamard flow) and oscillating-circulating droplets (mixed Hadamard-Dirichlet flow). Figure 4.5 shows calculated mass transfer results for six circulating droplet cases. Included is a matrix of runs at two Peclet numbers (10 and 100) for three different distribution coefficients (0.1, 1.0, and 10.0).

The overall rate of mass transfer increases as the distribution behavior of the transferring species becomes more favorable to the continuous phase (distribution coefficient = 0.1). This situation occurs because the concentration on the droplet side of the interface approaches zero which, in turn, increases the driving force for mass transfer into the continuous phase from the droplet. In addition, the calculated results indicate that increasing the Peclet number from 10 to 100 augments the overall rate of mass transfer. This occurs because convection is allowed to play a

Fig. 4.5. Computed mass transfer for six circulating droplet cases.

greater role in supplying material to the interface for subsequent transfer into the continuous phase. Figure 4.6 has plots of the calculated mass transfer data for an oscillating-circulating droplet at oscillation frequencies of 0, 10, 20, and 30 Hz. The model predicts that the overall rate of mass transfer is higher for an oscillating droplet than for a corresponding circulating, nonoscillating case. In the time required for 4.9% of the material to leave the nonoscillating droplet, mass transfer is 6.3% for the droplet oscillating at 10 Hz, 7.1% at 20 Hz, and 7.9% for the 30-Hz case. These numbers represent increases in mass transfer rates ranging from 29 to 57%. The model indicates that significant increases in the overall rate of mass transfer can be obtained at moderate oscillation frequencies and amplitudes. These results have been acquired using a low Peclet number (10) where mass

Fig. 4.6. Computed mass transfer for an oscillating-circulating droplet at various oscillation frequencies.

transfer rates are sensitive to changes in convective transport and, therefore, probably are near the upper limits which one might expect for increases in the overall rate via droplet oscillation. The effect should be less pronounced at higher Peclet numbers.

This supposition is qualitatively supported by earlier experimental work from this program, which reported an ~40% increase in the rate of mass transfer for droplets oscillating at 50 Hz (using a Peclet number of 50,000) and by recent experimental studies involving the transfer of fluorescent dyes into liquid droplets.

Mass transfer rates have been measured in single droplets by monitoring the change in fluorescence in the droplet, using a helium-cadmium laser for excitation and a photomultiplier tube to measure the intensity of fluorescence normal to the incident laser beam (Fig. 4.7). This system is capable of resolving nanogram quantities of fluorescent dye; hence, very precise mass transport experiments can be carried out. The flux of dye from the continuous phase (2-ethyl-1-hexanol) into an aqueous droplet is shown in Fig. 4.8, plotted as a function of oscillation frequency of the droplet. The flux of dye into the droplet increases as the frequency of oscillation increases, with the maximum rate of enhancement being on the order of 25% for the range of frequencies studied. The Peclet number for these experimental data is ~150,000; therefore, the results are in good agreement with the predicted

Fig. 4.7. Schematic of the apparatus used for mass transfer studies.

Fig. 4.8. Flux of dye from a continuous 2-ethyl-1-hexanol phase into an aqueous droplet as a function of oscillation frequency.

limits derived from the mass transfer model previously discussed.

Sorption and Extraction in the Critical Region

Work in this area results from a merger of two Chem Tech research programs; one program was concerned with solute-solvent interactions in supercritical extraction and the other with dynamics of adsorption. Since both programs were basically investigating molecular interactions in mixtures at extreme pressures, it was considered advantageous to focus both studies on similar systems.

Solvent-Solute Interactions in Supercritical Solvents. A theoretical framework, based on the statistical mechanical solution theory of Kirkwood and Buff, has been developed for describing isothermal solubility in a fluid of varying density.¹ Prior theories of this type have been restricted to isobaric conditions. The new expression is well-suited to modeling solubility in supercritical fluids where the density may vary widely with only small changes in pressure or temperature. The general expression for variation of the number density of a solute in an isothermal fluid at equilibrium with

1. H. D. Cochran, L. L. Lee, and D. M. Pfund, "Applications of the Kirkwood-Buff Theory of Solutions to Dilute Supercritical Mixtures," accepted for publication by *Fluid Phase Equilibria*.

another phase of fixed composition (e.g., for a solid or in bubble-point calculations) is as follows:

$$\frac{kT}{\rho_k} d\rho_k = (G_{kj} + \bar{v}_k) dP + \sum_{i \neq j}^c \rho_i (G_{ki} - G_{kj}) \bar{v}_i dP,$$

where

$G_{\alpha\beta}$ is the Kirkwood fluctuation integral for the interaction of any α - β pair:

$$G_{\alpha\beta} = \int_0^\infty [g_{\alpha\beta}(r) - 1] 4\pi r^2 dr,$$

in which

$g_{\alpha\beta}$ = the spatial pair correlation function for the α - β pair interaction,
 r = spatial separation between species α and species β .

Subscripts k , j , and i indicate the solute, the solvent, and the i th component in a c -component mixture, respectively, and the remaining variables are defined as follows:

ρ = number density,
 T = absolute temperature,
 P = pressure,
 $\bar{v}$ = partial molecular volume,
 k = Boltzmann constant.

In applications to dilute solutions, which are typical of most supercritical solubility data, the theory reduces to a simpler form:²

$$\ln(x_k) = \ln\left(\frac{p_k^0}{P}\right) - \frac{\bar{v}_k^s}{kT} (P - p_k^0) + Z_j^0 + \frac{1}{kT} \int_{p_k^0}^P G_{kj}^0 dP,$$

where

x_k = mole fraction of solute in the equilibrium mixture,
 p_k^0 = vapor pressure of pure solid solute,
 $\bar{v}_k^s$ = molecular volume of pure solid solute,
 Z_j^0 = compressibility factor of pure solvent,
 G_{kj}^0 = solute-solvent fluctuation integral at infinite dilution.

For useful applications of this expression, appropriate molecular solution approximations may be introduced to relate the term for solvent-solute interaction at infinite dilution, G_{kj}^0 , to that for the pure solvent, G_{jj}^0 . For example, expressions based on local composition approximations and excluded-volume approximations have been developed. When this is done, the solvent fluctuation integral may be calculated from an appropriate equation of state:

$$\rho_j^0 G_{jj}^0 = kTK_T - 1 = kT \left(\frac{\partial \rho_j^0}{\partial P} \right)_T,$$

where

K_T^0 = isothermal compressibility of pure solvent,
 G_{jj}^0 = Kirkwood fluctuation integral of pure solvent.

Very accurate equations of state are available for solubility in common supercritical solvents. The results of calculations with these models have been tested against published supercritical solubility data with satisfactory agreement.³ Figure 4.9 shows the results for naphthalene solubility in supercritical carbon dioxide calculated using the excluded-volume approximation with temperature-dependent parameters. Figure 4.10 shows results for the same system calculated using the local composition approximation; in this case all temperature dependence is contained within the local composition model.

Published data for testing theoretical models of solubility in supercritical fluids are largely restricted to systems in which solid, organic solutes of low volatility are dissolved in common gases that have critical temperatures near ambient—carbon dioxide and ethene being the most commonly studied. To broaden the available data base and explore for possible applications related to DOE's mission, we have initiated measurements of the solubility of volatile transition-metal fluorides, such as MoF_6 and UF_6 , in supercritical CF_4 . The apparatus for these experiments is shown schematically in Fig. 4.11. Subambient temperatures ($T_c = 227.6$ K) are maintained, while the compressed

2. H. D. Cochran and L. L. Lee, "General Behavior of Dilute Binary Solutions," accepted for publication by *AIChE Journal*.

3. D. M. Pfund, L. L. Lee, and H. D. Cochran, "Application of the Kirkwood-Buff Theory of Solutions to Dilute Supercritical Mixtures. II. The Excluded-Volume and Local-Composition Models," submitted to *Fluid Phase Equilibria*.

Fig. 4.9. Solubility of naphthalene in supercritical carbon dioxide. Curved lines are predictions from the excluded-volume model with temperature-dependent parameters.

Fig. 4.10. Solubility of naphthalene in supercritical carbon dioxide. Curved lines are predictions of the local-composition model.

CF_4 ($P_c = 3.74 \text{ MPa}$) passes through a bed of solid solute in the extraction cell. Initial results for MoF_6 solubility in supercritical CF_4 are shown in Fig. 4.12. Although there are known errors in the measured solubility at the high and low concentration limits, these data are in reasonable agreement with predictions from the excluded-volume model.

Sorption Studies. Chemical separations techniques such as ion exchange, physical adsorption, solvent extraction, continuous chromatography, and sorption have long been studied by Chem Tech with the aim of improving our basic understanding of these processes, and the results of this work often have been successfully applied in industry. Sorption is not only an important separation pro-

cess on its own but is also a vital step in many other separations and chemical processes. Virtually no separation can take place at an interface between any two phases without sorption as the first and final steps, regardless of the intervening mechanisms.

The purpose of the Chem Tech sorption studies is to relate the dynamics of sorption (e.g., adsorption rate, desorption rate, and surface diffusivity) and the statics of sorption (e.g., surface residence times, separation factors, and spreading pressures) to the intermolecular forces which control these effects. Theoretical, as well as experimental, evaluations of the various parameters which affect adsorption are being done.

During this period, the work was focused in the following two areas: (1) the development of statistical-mechanical equations for the adsorption of pure components and mixtures in cavities such as those found in molecular sieve adsorbents, and (2) the design and construction of experimental equipment which will enable us to measure quantities related to the dynamics of sorption, with special emphasis on multicomponent systems.

The model statistical-mechanical equations for pure component adsorption in microporous adsorbents have been developed and applied to numerous systems. These have been found to be reliable, showing consistently better results than previously existing theories and giving physically meaningful values for quantities such as collision diameters and force constants. An extension of the theory to predict mixture isotherms from pure component parameters has been accomplished recently. Future work will apply this same approach to adsorption on flat surfaces. Because the theory yields microscopic properties, these properties can be used with other theories to predict dynamic effects of adsorption.

In our experimental work, we have initiated the development of new techniques which will allow the direct measurement of sorption dynamics. A piezoelectric microbalance assembled for this purpose utilizes the frequency of vibration of a quartz crystal to determine the adsorptive loading of the crystal, since the frequency shift of surface acoustic waves in quartz is directly proportional to the mass applied to the surface. It has further been found that such quartz crystal sensors can be used for both gases and liquids. A study of both the statics and dynamics involved in adsorption of gases, gas mixtures, and liquid mixtures on the

Fig. 4.11. Simplified schematic of equipment used for measuring solubility of volatile transition-metal fluorides in supercritical CF_4 .

Fig. 4.12. Measured solubility of MoF_6 in CF_4 at 243 K. Curve shows predictions from the excluded-volume model.

bare crystal surfaces would be extremely useful. However, a study of adsorption in or on metal films applied to quartz crystals could be even more valuable.

Sorption Pumping and Transfer Operations Using Deep Beds of Sorbents

Cryosorption of gases and vapors in beds of sorbent materials is important in several existing processes and has been proposed for use in the safe storage of fuel gases and in the high-vacuum fusion energy operations. The fusion energy applications will require clean roughing pumps to evacuate chambers with volumes up to 10^4 m³, and large cryosorption pumps that use molecular sieve beds at liquid-nitrogen temperatures are under consideration for this service. A more-detailed understanding of the heat- and mass-transfer processes associated with deep beds of sorbents will be required to design and model the behavior of such large units.

For the Chem Tech research work in this area, we simulate deep beds of sorbent with a cylindrical

Fig. 4.13. Apparatus used for vacuum cryosorption studies of nitrogen and carbon dioxide.

sorbent bed at high-vacuum conditions, immersed in a coolant such as liquid nitrogen.⁴ Details of the apparatus are shown in Fig. 4.13. A constant feed of gas is admitted to the space above the bed, and the pressure is monitored at several locations. The experiments have been performed with 4A molecular sieves, using nitrogen and carbon dioxide as the adsorbed species.

Based on more recent research, a new theoretical model has been developed which describes adsorption of carbon dioxide and nitrogen on 4A zeolite molecular sieves at cryogenic temperatures.⁵ The model includes a fluid-phase mass balance, representing the dynamics of gas in the bed, and one-

dimensional diffusion, describing adsorption in the solid. Zeolite crystals are modeled as mono-dispersed spheres. The concentration dependence of the sorbate's diffusivity in both the gaseous and solid phases is incorporated in the model. The Langmuir isotherm is used to represent equilibrium concentrations at the gas-sorbent interface. Numerical solution of the nonlinear, parabolic, partial differential equations which result from the physical model is accomplished by orthogonal collocation in conjunction with an ordinary differential equation integrator suitable for stiff equations. The method of solution is approximately two orders of magnitude faster than the previous finite difference solutions. Stability problems have been eliminated by this approach.

Experimental observations obtained by Crabb et al.⁴ were compared with the theoretical results. As shown in Fig. 4.14, good agreement with experimental results for carbon dioxide cryosorption was obtained, indicating that the primary diffusional resistance occurs in the microporous zeolite crystals

4. K. S. Crabb, J. J. Perona, C. H. Byers, and J. S. Watson, "Vacuum Sorption Pumping Studies with Pure Gases on Molecular Sieves," *AIChE J.* 32, 255 (1986).

5. J. M. Prazniak and C. H. Byers, *A Modeling Study of Vacuum Sorption Characteristics of Carbon Dioxide on 4A Zeolite Molecular Sieves*, ORNL/TM-10327, in publication (December 1986).

Fig. 4.14. Calculated and experimental values for cryosorption of carbon dioxide on zeolite molecular sieves.

rather than in the bed interstices or the macropores formed by the clay binder used to pelletize the crystals. In contrast, the relative agreement of theoretical and experimental results for nitrogen cryosorption supports the conclusion reached earlier by Crabb et al., that the controlling diffusional resistance in nitrogen cryosorption is in the bed interstices and micropores.

The new sorption model allows us to efficiently model deep-bed sorption experiments for a broad range of materials and for such applications as the fusion energy sorption pumping and safe storage of fuels. The computer algorithm's usefulness for a wide range of fixed-bed modeling problems is currently under investigation.

Phase Equilibria of Solutions in the Critical Region

In the production of synthetic natural gas, the unreacted feed components and some of the products must be removed before the gas can enter the natural gas pipeline system. A new concept for dealing with this problem is shown in Fig. 4.15. Liquid carbon dioxide near its critical point is used as an absorption medium. In the first of two adsorption phases, the excess hydrogen, carbon

Fig. 4.15. Schematic process diagrams for the recovery of methane from a syngas gasification reactor.

monoxide, and carbon dioxide are recycled to the reactor vessel; the second phase removes the heavier materials such as hydrogen sulfide, carbonyl sulfide, and the heavier hydrocarbons and releases methane into the gas pipeline at the appropriate pressure. Clearly this is more energy efficient than the conventional cryogenic process, but the extent of the savings is not fully known.

It is difficult to assess the influence of the various multicomponent phase equilibria of the light gas systems upon the ASPEN simulation model that has been prepared to optimize operation of the system prior to full-scale implementation. Therefore, a Chem Tech experimental program was initiated to resolve problems in predicting multicomponent equilibria in systems which are primarily carbon dioxide and methane, with small amounts of hydrogen, carbon monoxide, hydrogen sulfide, and carbonyl sulfide at near-critical conditions. The experimental data gathered in this program will be particularly valuable because equations of state perform poorly in predictions for the critical region.

A high-pressure, vapor-liquid equilibrium apparatus built for this program (Fig. 4.16) contains a central high-pressure cell attached to a magnetically operated recirculation pump. Composition data are collected using a gas chromatograph. The construction and testing of the apparatus were completed during the period of this report.

Data have been collected for several binary gas-liquid systems. Typical of these are the data shown in Fig. 4.17. The ternary $\text{CO}_2\text{-CH}_4\text{-H}_2\text{S}$ system has also been studied. Of several equations of state

Fig. 4.17. Data for phase equilibria of the gas-liquid methane-hydrogen sulfide system.

considered for prediction of the data, the Redlich-Kwong-Soave and the Peng-Robinson equations were selected to fit the data. Both equations showed a good correlation capability; in addition, our data show excellent agreement with reliable literature values. Therefore, we are proceeding with our system simulation work with a considerable degree of confidence.

4.2 MATERIALS SCIENCES

The DOE Office of Basic Energy Sciences (BES), through the Division of Materials Sciences,

Fig. 4.16. Schematic of the high-pressure, vapor-liquid apparatus used for equilibrium studies in the critical region.

supports Chem Tech research programs concerned with thermodynamics and sensitivity analyses of energy-related systems, as well as basic chemical engineering studies.

Thermodynamics of Energy-Related Materials

The objectives of this work are the determination and modeling of phase equilibria and other thermochemical data important to energy-producing systems. Current emphasis is on the measurement and application of such data for the actinide oxides used as nuclear fuels, but the methodology is applicable to any oxide solid solution. Relevant subsystems of the elements U, Pu, O, and specific fission products, particularly lanthanides, are currently being studied under conditions generally not previously investigated. Our own adaptations of chemical and mathematical models from the literature are being used to represent the ~5000 values of chemical potential and nonstoichiometry reported in the literature. These efforts provide a previously unavailable, generalized, chemical thermodynamic description of the actinide-lanthanide dioxide solutions.

A miscibility gap in the $\langle \text{PuO}_{2-x} \rangle^*$ phase at 930 K has been incorporated into the previously published chemical thermodynamic representation.⁶ This improvement was published as a letter to the editor.⁷

The $\langle \text{CeO}_{2-x} \rangle$ phase is nonstoichiometric over the composition range $0 < x < 0.3$, and there are ~1000 values of x in the literature for known temperatures and chemical potentials of oxygen. The entire $\langle \text{CeO}_{2-x} \rangle$ data base for the dependence of the nonstoichiometry, x , on temperature and chemical potential of oxygen (oxygen potential) was retrieved from the literature and represented by a thermodynamic method.⁸ The method resulted in partial molal Gibbs free energy quantities that are useful for any thermodynamic calculation involving $\langle \text{CeO}_{2-x} \rangle$. It also accurately represents, for the

first time, the complex behavior reflected in the experimental data, including the low-temperature miscibility gap. The method appears to be generally applicable to many very nonstoichiometric phases.

The final assessment of this study was for ~850 data points for the solutions of lanthanide oxides in uranium dioxides. No previous assessment or thermodynamic representation of these data had been done. The literature data base for the dependence of the nonstoichiometry, x , on temperature and on the chemical potential of oxygen in $\langle \text{U}_{1-x}\text{Ce}_x\text{O}_{2+x} \rangle$ and $\langle \text{U}_{1-x}\text{Ln}_x\text{O}_{2+x} \rangle$ was retrieved and represented by a thermodynamic method;⁹ the method used reproduces the results from the experimental measurements and also gives partial molal Gibbs free energy quantities that are useful for any thermodynamic calculation involving these nonstoichiometric phases. The behavior of each of these systems is also compared with that for $\langle \text{U}_{1-x}\text{Pu}_x\text{O}_{2+x} \rangle$.

A thermodynamic analysis of the chemical vapor deposition of the desired composite ceramic $\langle \text{Si}_3\text{N}_4 \rangle$ - $\langle \text{BN} \rangle$ was also performed in this study, using a general computer program for the calculation of heterogeneous equilibria.¹⁰ The assumed reactants were the gaseous species H_2SiCl_2 , PCl_3 , and NH_3 . The equilibrium conditions for the deposition of condensed phases in the B-Si-N system were determined as a function of the temperature, total system pressure, and gram-atomic reactant gas fractions $\text{B}/(\text{Si} + \text{B})$ and $\text{N}/(\text{Cl} + \text{N})$. The phase assemblage, $\langle \text{Si}_3\text{N}_4 \rangle$ - $\langle \text{BN} \rangle$, was found to be stable over a large region. Predominant gaseous species at equilibrium were HCl , N_2 , H_2 , and the silicon chlorides. Deposition efficiencies at equilibrium for $\langle \text{Si}_3\text{N}_4 \rangle$ and $\langle \text{BN} \rangle$ were high, particularly in the presence of excess gaseous NH_3 and at temperatures < 1600 K.

* $\langle \rangle$ denotes solid phase.

6. T. M. Besmann and T. B. Lindemer, "Chemical Thermodynamic Representation of $\langle \text{PuO}_{2-x} \rangle$," *J. Nucl. Mater.* 130, 489-504 (1985).

7. T. M. Besmann and T. B. Lindemer, "Improvement in the Chemical Thermodynamic Representation of $\langle \text{PuO}_{2-x} \rangle$ and $\langle \text{U}_{1-x}\text{Pu}_x\text{O}_{2+x} \rangle$," *J. Nucl. Mater.* 137, 292-93 (1986).

8. T. B. Lindemer, "Chemical Thermodynamic Representation of Very Nonstoichiometric Phases: $\langle \text{CeO}_{2-x} \rangle$," *CALPHAD* 10, 129-36 (1986).

9. T. B. Lindemer and J. Brynestad, "Review and Chemical Thermodynamic Representation of $\langle \text{U}_{1-x}\text{Ce}_x\text{O}_{2+x} \rangle$ and $\langle \text{U}_{1-x}\text{Ln}_x\text{O}_{2+x} \rangle$; Ln = Y, La, Nd, Gd," *J. Am. Ceram. Soc.* 69, 867-76 (1986).

10. T. M. Besmann, "Thermodynamic Analysis of the Chemical Vapor Deposition of Composite $\langle \text{Si}_3\text{N}_4 \rangle$ - $\langle \text{BN} \rangle$ Coatings," *J. Am. Ceram. Soc.* 69, 69-74 (1986).

Nucleation, Growth, and Transport Phenomena

Two interdependent research programs in Chem Tech utilize laser-light-scattering experimental techniques to investigate the phenomena of homogeneous precipitation and particle growth. The first is a series of fundamental studies of homogeneous nucleation and growth in systems which are of interest as precursor materials in ultrafine processing for a new generation of ceramic materials. The second area of interest is the measurement and theoretical interpretation of transport properties of fluids in the critical region. The transport properties, such as viscosity and diffusion coefficient, provide important clues to the behavior of the fluid media in which the crystallization occurs. The latter study has achieved maturity, and our research emphasis is shifting to the precipitation work.

Particle Growth Kinetics in the Homogeneous Precipitation of Silica and Titania by Metal Alkoxide Hydrolysis. A central issue in the development of a new generation of ceramics for use under extreme conditions has been gaining an understanding of the chemical factors which control the generation of powders suitable for tailoring superior properties. The base materials are usually precipitated metal oxides which are either drawn from natural sources or made synthetically. Synthetic oxides generally produce better, more reproducible properties in the resulting ceramic. One of the most important factors associated with success in this field is the production of monodispersed powders in the submicron range. Equally important is the ability to control and characterize the material as it is produced.

This program has included several series of fundamental investigations of homogeneous nucleation and growth in systems of ceramic precursor materials. The hydrolysis of alkoxides and the subsequent condensation reactions result in a homogeneous precipitation of oxide powders, such as silica and titania. These are studied in a laser-light-scattering facility using a thermally controlled quartz cell as a crystallization vessel. Rapid, noninvasive monitoring of particle growth in the 0.01- to 1- μm range is routinely achieved.

A study of the particle growth and powder characteristics of titania and silica indicates that a major contributing factor to the ultimate particle size and morphology is the nature of the alcohol

solvent used as a medium for the reaction. Titania particles were synthesized by titanium ethoxide hydrolysis in ethanol, 1-butanol, and *t*-butanol solvents. The hydrolysis reaction is rapid for this compound, and particle growth is controlled by the rate of precipitation of the hydrolysis product. Particle growth from 0.015 to 1.5 μm is monitored by dynamic laser-light scattering. Figures 4.18 and 4.19 show particle growth kinetics for reactions in ethanol and 1-butanol during the period when the concentration of soluble titania is essentially constant. The linear growth (i.e., constant growth rate) is characteristic of controlled surface-nucleation growth. Particle growth kinetics in *t*-butanol is illustrated in Fig. 4.20. This plot shows an increase in the rate of growth with time, which is characteristic of uncontrolled nucleation [i.e., the rate of surface nucleation (particle growth) is slower than the rate of particle formation]. Electron microscopy showed that the particles produced in ethanol and 1-butanol were monodisperse and spherical. Particles grown in *t*-butanol were of random shapes and sizes.

The synthesis of silica by the hydrolysis of tetraethylorthosilicate (TEOS) in ethanol, the two propanols, three butanols, and *t*-amyl alcohol has also been investigated. The hydrolysis reaction of TEOS is slow in comparison to the condensation reaction, allowing the use of gas chromatography to monitor the rate of hydrolysis. Results for the

Fig. 4.18. Particle growth kinetics for titanium ethoxide (TIE) hydrolysis in ethanol.

Fig. 4.19. Particle growth kinetics for titanium ethoxide (TIE) in 1-butanol.

Fig. 4.21. Solvent effects on the hydrolysis rate of TEOS (tetraethylorthosilicate).

Fig. 4.20. Particle growth kinetics for titanium ethoxide (TIE) in *t*-butanol.

Fig. 4.22. Solvent effects on total soluble silica kinetics: TEOS (tetraethylorthosilicate) hydrolysis in various alcohols.

alcohol systems indicate that the hydrolysis reaction is strongly influenced by both the degree of hydrogen bonding between water and the solvent and by steric factors. Figures 4.21 and 4.22 illustrate the effect of solvent on the hydrolysis rate and on the condensation rate (which is determined by the rate of decrease in the total soluble silica concentration). The nucleation rate, which controls the ultimate particle size, is most strongly influenced by physical factors, principally viscosity. Figure 4.23 shows electron micrographs of monodisperse silica particles synthesized in 2-butanol. Initial tests have also been conducted on the synthesis of zirconia particles by the hydrolysis of

zirconium-*n*-butoxide in 1-butanol. Monodisperse spherical particles were produced.

Transport Properties of Fluids at Extreme Conditions. This program applies dynamic laser-light scattering to the study of high-temperature and/or high-pressure fluid properties.¹¹ Small sample volumes (<5 mL) are required, which simplified the temperature and pressure control. Light-scattering techniques enable us to relate the

11. D. F. Williams and C. H. Byers, "Determination of High-Temperature Fluid Viscosity Using Dynamic Light Scattering," *J. Phys. Chem.* **90** (11), 2534 (1986).

Fig. 4.23. Electron micrograph of monodisperse silica particles synthesized in 2-butanol.

dynamics of the molecular or colloidal species being characterized to the transport properties of the fluid. The mobility of the colloidal species is characterized as a diffusion coefficient, while an application of the Stokes-Einstein relationship to the model colloid system allows calculation of the fluid's viscosity. Our present work focuses on measuring the viscosity of supercooled liquids and of nonideal, binary, organic mixtures at high temperatures, using model colloids as the suspended scattering species. Stabilized silica particles ($0.1\text{-}\mu\text{m}$) are used as model colloids for the high-temperature viscosity experiments with nonpolar fluids. Work has also continued on measuring the viscosities of mixtures near the critical point. Early experiments in this series verified the method, using simple hydrocarbons such as pentane, hex-

ane, and heptane and their mixtures. These studies also showed that data for the viscosities of simple pure components can be accurately predicted for the near-critical region, using corresponding-states theory. However, viscosities of more complex materials such as acetone and phenol are not as well predicted.

The viscosities of ethanol-heptane mixtures were measured experimentally at temperatures ranging from ambient to 230°C . Standard polystyrene spheres were used as model colloids for determining the viscosity of supercooled *t*-butanol. The data from light-scattering methods compare favorably with values acquired using capillary viscometry. This is important because it shows that our results are not an artifact of the method of measurement. Two Andrade (a two-parameter model) curves are

required to fit the *t*-butanol data in Fig. 4.24, suggesting a significant increase of the intermolecular forces in the melting-point region.

Capillary viscometry was used to determine the viscosity of supercooled phenol. Two Andrade curves (one above and one below the freezing point) cross near the freezing point of phenol (Fig. 4.25).

Fig. 4.24 Viscosity data for pure *t*-butanol in the melting point region, showing correlations made using two Andrade relationships.

Fig. 4.25. Correlation of pure phenol viscosity data using a three-parameter Antoine relationship.

5. BIOTECHNOLOGY RESEARCH

Research and development in biotechnology is carried out in four major areas: (1) applied biological research in support of processing concepts; (2) bioengineering research that provides the bridge between the more fundamental work and processing systems; (3) bioprocessing research in which the technical feasibility of specific processing concepts is established; and (4) research on bioanalytical technology.

5.1 BIOLOGICAL RESEARCH

Light-Induced Water Splitting by Biological Systems

Photosynthesis research in the Chemical Technology Division has focused on understanding the physicochemical aspects of photosynthesis, with specific application to developing a biotechnological process for production of gaseous fuels from renewable inorganic materials. Two factors contribute to making photosynthesis a key area of research. First, photosynthesis is a self-replicating process that is capable of synthesizing energy-rich molecules from abundant inorganic materials, including atmospheric carbon dioxide, which serves as the *sole* carbon source. Second, we have shown that, under appropriate physiological conditions, certain freshwater and marine green algae, as well as the reconstituted *in vitro* system composed of isolated chloroplasts, ferredoxin, and hydrogenase, are capable of splitting water to molecular hydrogen and oxygen in a sustained steady-state reaction. In these systems, the gaseous-fuel-producing reaction can be driven by light throughout the visible portion of the solar emission spectrum, including the long wavelength (red), 700-nm region. No external energy sources are required. These two facts, combined with previous and recent experimental results, indicate that photosynthetic water splitting is a promising research area for the production of gaseous fuels from inorganic resources.

A significant advance in microalgal water splitting for gaseous fuel synthesis has been made during the past year. In the linear, low-intensity region of the light saturation curve, we have measured equivalent solar conversion efficiencies of $\sim 10\%$. This efficiency is, by far, the highest value ever reported for a biological system. Working with a thin film (<50 monolayers) of the unicellular green algae *Chlamydomonas reinhardtii* and *Scenedesmus* D₃, equivalent solar conversion efficiencies in the range of 3 to 12% have been measured, as summarized in Table 5.1. These values were computed from the net amount of visible light absorbed by the algae entrapped on the filter paper and the fact that the visible region constitutes approximately half of the power radiated in the solar emission spectrum. The key advance realized in this work is the recognition of the importance of the physiological state of the algae in terms of film thickness and usable light energy.

Submicron Bioelectronic Materials and Structures

The photochemical apparatus of green plant photosynthesis is a light-activated molecular electronic device. The photovoltages that are generated and photocurrents that flow in living tissue occur in a plant organelle called the chloroplast. The electronic components of the chloroplast are embedded in the inner membranes, which are composed of hydrocarbons. The molecular-electronic components are made up of proteins containing functional groups that are the active centers associated with electron flow.

Unlike metals and semiconductors, whose electronic properties are determined by bulk properties of matter, electron flow in the photosynthetic electron-transport chain takes place between discrete molecular entities. This novel method of electron flow and its occurrence in a thin membrane provide for a highly versatile system of charge separation and energy transduction. One example

Table 5.1. Energy conversion efficiencies of green algae for hydrogen and oxygen production

Exp. No.	Alga	Irradiation interval ^a	Efficiency (PAR) (%) ^b	Equiv. solar eff. (%) ^c
1	<i>Scenedesmus</i> D ₁	1	16	8
		2	23	11.5
2	<i>C. reinhardtii</i> (sup)	1	13	6.5
		2	16	8
		3	18	9
		4	19	9.5
		5	21	10.5
		6	21	10.5
		7	18	9
3	<i>C. reinhardtii</i> (UTEX 90)	1	6	3
		2	8	4
		3	8	4
4	<i>C. moewusii</i>	1	24	12
		2	22	11
		3	18	9

^aThe irradiation intervals consisted of sequential cycles of 3 h ON-3 h OFF.

^bConversion efficiency based on absorbed photosynthetically active radiation.

^cColumn 2 divided by 2, assuming the photosynthetically active radiation comprises half of the power in the solar emission spectrum.

of this versatility would be the differential affinity of some discreet membrane components for protons—depending on the presence or absence of an electron on the molecule. The ability to bind and release protons by plastoquinone results in the coupled translocation of protons across the photosynthetic membrane with the flow of electrons within the membrane. Under the influence of light, the flow of electric current in photosynthetic membranes occurs in two mutually orthogonal directions and with different charge carriers. The two currents are normally linked by the molecular nature of the particular electron transport chain. It is possible, however, to uncouple the electron flow from the proton translocation.

In normal photosynthesis, electrons emerge at the surface of the membrane and are used to reduce the coenzyme nicotinamide-adenine dinucleotide phosphate (NADP⁺) to NADPH, the electron carrier for the Calvin cycle in which carbon dioxide is enzymatically reduced to a carbon dioxide fixation compound. The unique feature of the research described in this section is the direct electrical contact with emergent electrons and the utilization of these electrons in two ways: (1) for

the electrosynthesis of molecular hydrogen and (2) for the flow of electrical current in an external circuit.

Colloidal platinum was precipitated directly onto photosynthetic membranes in an aqueous suspension. The resulting chloroplast-colloidal platinum composition was then entrapped on filter paper. As indicated in Fig. 5.1, this moistened material was capable of sustained simultaneous photoevolution of hydrogen and oxygen when irradiated with visible light. Since no electron relay was added to the system and the overall reactions occurred in an immobilized matrix, we concluded that the precipitated colloidal platinum directly contacts the reducing end of photosystem I in such a way that electron flow occurs across the biological membrane-metal-colloid interface with preservation of charge continuity and catalytic activity. In addition to the special photocatalytic properties of platinumized chloroplasts, the specific ionic species used to prepare the material yield information on the physicochemical properties of the photosystem I reduction site on the thylakoid membrane surface. The data in Fig. 5.1 were obtained by precipitating platinum from the hexachloroplatinate(IV) ion,

Fig. 5.1. Data for the simultaneous photoproduction of hydrogen and oxygen by maintained platinized chloroplasts entrapped on filter paper.

$[\text{Pt}(\text{Cl})_6]^{2-}$. Platinized chloroplasts that were prepared by precipitating platinum from the tetraamineplatinum(II) ion, $[\text{Pt}(\text{NH}_3)_4]^{2+}$, resulted in no hydrogen activity and only a transient oxygen gush. The presence of insoluble platinum in the filter paper-entrapped composition was determined by X-ray fluorescence analysis.

The time profiles of photoactivity are presented in Fig. 5.1 for three light-dark cycles of 2 h each. Figure 5.1 shows that the first irradiation period has a time profile that is qualitatively different from the profiles of the subsequent two periods. In the first cycle, the oxygen profile underwent a transient gush (peaking at 7 μmol of O_2 per hour) before settling down to steady state. The hydrogen rate climbed monotonically to steady state. The time required to reach 50% of steady state in the first cycle was ~ 25 min, whereas the corresponding times for the second and third cycles were 5 to 8 min, respectively.

Electrical contact with the platinized chloroplasts was achieved by contacting the surface of the filter paper-entrapped material with a fine-mesh platinum gauze. A fine-mesh silver gauze, placed on the opposite side of the filter paper, was used as the counter electrode. All of the components were held in close proximity in a Lucite sandwich-like assembly with nylon compression screws, as indicated in Fig. 5.2. Platinum lead wires, which were connected to the input of an appropriate digital multimeter for current and voltage measurements, were used to make electrical

contact with the platinum and silver gauzes. All electrical and gas evolution measurements were performed in a humidified helium atmosphere.

The simultaneous photoevolution of hydrogen and oxygen strongly suggests that the chemically precipitated platinum physically contacts the reducing side of photosystem I on the thylakoid membrane surface. Since considerably more platinum was precipitated than is necessary for catalytic activity, electrical contact with the photosynthetic electron transport chain can be achieved by pressing the platinized chloroplast-filter paper composition against a gauze electrode. The data of Fig. 5.3, showing the short-circuit photocurrent for consecutive ON-OFF light cycles, indicate that this is indeed possible.

Several noteworthy aspects of the data in Fig. 5.3 are evident. First, the platinized chloroplasts that give rise to the photocurrent are unoriented and presumably not all electrically contiguous. This structure implies that only a fraction of the functional chloroplasts actually contribute to the observed photocurrent. Although the chloroplasts are unoriented, the platinum is precipitated on the external surface of the thylakoid membranes. This external surface also corresponds to the reducing end of photosystem I. The network of colloidal platinum, platinized chloroplasts, and platinum gauze forms an isopotential surface that gives rise to the directionality of current shown in Fig. 5.3.

Note also that the sign of current flow indicates that electrons flow from the platinum electrode to

Fig. 5.2. Schematic drawing of a compression assembly for measuring photoeffects in platinized chloroplasts.

Fig. 5.3. Plots of the short-circuited photocurrent generated by the entrapped platinized chloroplasts contained in the compression assembly of Fig. 5.2.

the silver/silver chloride electrode through the external circuit. This directionality of current flow agrees with the generally accepted vectorial model of photosynthesis, in which the external surface of the thylakoid membrane is driven to negative potentials with respect to the inner thylakoid surface under the influence of light.

Third, the current-versus-time profile of the first cycle shown in Fig. 5.3 is qualitatively different from those of the second and third cycles. The relatively slow increase of current in the first cycle is interpreted as meaning that a major fraction of the electric current is performing electrochemical processes. When the light is turned on the second and third times, the current-versus-time profile shows two distinct features: (1) a capacitive, interfacial-charging component and (2) a long-term, steady-state, direct current (DC) component. The presence of this long-term DC component demonstrates electrical contact with the electron transport chain of photosynthesis.

Recent research focusing on a new method for chemically modifying the surface of thylakoid membranes and making electrical contact with the electron transport chain of photosynthesis has been described in this section. The primary light reaction occurs in a molecular structure that is several nanometers in size. Moreover, since the characteristic switching time of the primary process of photosynthesis is in the picosecond time scale, it follows that discreet molecular devices based on direct electrical contact with photosynthetic reaction centers could offer an interesting area of research for the development of very fast molecular electro-optical devices.

Enzyme Interactions with Solid Surfaces

The interaction of an enzyme with a solid surface, which is also a substrate for that enzyme, is important because the enzyme-catalyzed reaction takes place in a heterogeneous medium. In order for the catalysis to proceed, the enzyme must first adsorb onto the surface of the substrate followed by the reaction itself. An understanding of the adsorption mechanism and the chemical nature of attachment of enzyme to insoluble substrate is of fundamental importance in determining the mechanism of catalysis.

The enzyme that we have chosen as a model for studying this phenomenon is cellulase, which catalyzes the hydrolysis of insoluble cellulose to glucose. Cellulase, however, is not a single enzyme but is composed of two major components that are required to bring about hydrolysis—namely, cellobiohydrolases and endoglucanases. Cellulases can be of bacterial, fungal, or plant origin; that used in our studies is of fungal origin. Cellulase from *Trichoderma reesei* hydrolyzes insoluble cellulose, whereas that from *Aspergillus niger* has little reactivity toward this substrate. Both cellulases, however, have similar activity toward soluble cellulosic substrates such as carboxymethylcellulose. Our approach to these studies focuses on determining the reason *Aspergillus niger* cellulase does not hydrolyze insoluble cellulose. Possible explanations for this could be that (1) the enzyme does not adsorb to cellulose and (2) a critical component is inactive or absent in the cellulase complex.

Purified cellobiohydrolase isoenzymes from *T. reesei* (CBH I and II) and endoglucanases (EG I and II) have been obtained from Dr. Sharon Shoemaker of Genencor, San Francisco. Both of these components adsorb to cellulose. Scatchard analysis of the equilibrium binding data for EG I (Fig. 5.4) reveals that the adsorption is biphasic. Native cellulose is mostly crystalline with a much smaller amorphous region. The region of the curve having a K_d (affinity constant) value of 4.83 L/g indicates a high affinity for EG I; the total number of these high-affinity binding sites is 0.273 mg/g cellulose. On the other hand, the region of the curve having a $K_d = 0.034$ L/g indicates a much lower affinity but a total number of binding sites of 4.77 mg/g cellulose. These data correspond to the binding of EG I to the amorphous and crystalline regions of cellulose, respectively. In order to effect the hydrolysis of native cellulose, binding of the cellulase components to the crystalline region must occur.

The endoglucanase activity of *A. niger* cellulase can be separated into at least three components by affinity chromatography of this cellulase on concanavalin A-Sepharose (CAS), which binds glycoproteins containing glucose and/or mannose residues (Fig. 5.5). Fractions 7 to 20, 44 to 52, and 53 to 61 containing the endoglucanase activity were pooled and are hereafter referred to as endoglucanase (EG) A, B, and C, respectively.

ORNL DWG 87-332R

Fig. 5.4. Affinity chromatography of *A. niger* endoglucanase.

ORNL DWG 86-527 R

Fig. 5.5. Scatchard plot for EG-1 from *Trichoderma reesei*.

The data in Table 5.2 indicate that EG B and EG C are glycoproteins because they bind to CAS. They can be eluted by using 0.3 to 0.7 *M* mannose. Although they have not been purified to homogeneity, it is noteworthy that a high percentage of carbohydrate is associated with these enzyme fractions. Thus, the EG B and EG C fractions possess 57% and 48% carbohydrate, respectively, presumably containing mannose. Interestingly, the EG A fraction that does not bind to CAS also appears to contain a large concentration of carbohydrate compared with the protein concentration. The nature of the carbohydrate of EG A is unknown, but it does not contain predominantly mannose or glucose; otherwise, it would have bound to CAS.

Table 5.2. Summary of the separation of endoglucanase fractions A, B, and C after affinity chromatography on CAS

Parameter	Endoglucanase fraction		
	A	B	C
Volume, mL	36.5	28.0	24.0
Protein, mg	0.25	0.67	0.24
Carbohydrate, mg	2.3	0.9	0.22
Activity			
Total units	102	168	41
Units/mg protein	408	251	171

These three endoglucanases did not adsorb on microcrystalline cellulose (Avicel) to any great extent. It is interesting to speculate that a high carbohydrate content may hinder the adsorption of endoglucanase on the cellulose surface and reduce the ability of a cellulase to hydrolyze an insoluble cellulosic substrate. This possibility is currently under investigation. The interaction of cellulase

with a solid surface such as CAS can be used, therefore, in the purification of cellulase components. During the purification of the β -glucosidase component of *A. niger* cellulase by affinity chromatography on CAS, an unusual phenomenon was observed. Mannose (up to 1.08 *M*) was ineffective as an eluent of this enzyme from CAS, as were several other sugars and their derivatives, including 0.5 *M* glucose. Also, washing the CAS: β -glucosidase complex with buffer at pH 3.5 in the absence of $MnCl_2$ and $CaCl_2$ (required to preserve the binding activity of concanavalin A below pH 5.0) did not result in elution of this enzyme. Therefore, affinity chromatography (using CAS) of *A. niger* β -glucosidase should not be used in the purification of this enzyme. The reason it is extremely difficult to elute *A. niger* β -glucosidase from CAS using the usual conditions for elution of glycoproteins from this support is unknown. On the contrary, CAS provides an excellent support material for the preparation of a highly active, immobilized form of this enzyme that will not readily leak from the support and can be used for the continuous hydrolysis of a cellobiose solution.

Finally, the interaction of cellulase components with solid surfaces other than their substrate has been tested to determine whether such surfaces can be used for the recovery of the enzyme from a cellulosic hydrolysate.

The adsorption of *T. reesei* cellulase protein to DEAE-, CM-, cibracon blue-, and concanavalin A-Kieselguhr-based supports (protein adsorbents termed Macrosorb) has been measured. The DEAE- and CM-Macrosorb possessed the greatest capacity for cellulase protein that was unaffected by 0.5 *M* glucose and minimally affected by 10 vol % ethanol (Table 5.3).

Table 5.3. Adsorption of cellulase protein by four Macrosorb preparations^a

Additive	DEAE-Macrosorb	CM-Macrosorb	Cibracon blue-Macrosorb	Concanavalin A-Macrosorb
None	46	47	28	29
Glucose, 0.5 <i>M</i>	48	47	17	21
Ethanol, 10 vol %	39	41	10	14

^aValues given are the percentages of cellulase protein (i.e., 5.4 mg) initially contacted with the support that is adsorbed to the Macrosorb preparation. Adsorption was conducted in 50 mM sodium acetate buffer, pH 5.0.

Adsorption of protein to DEAE-Macrosorb was measured at pH 3.5, 5.0, 6.0, and 7.0. At pH 3.5 and 7.0, 25 and 76% of the protein contacted with the adsorbent was bound, respectively. Cellulase protein that adsorbed on DEAE-Macrosorb at pH 7.0 was eluted by shaking the support in 10 mM sodium acetate buffer, pH 3.5. The eluted protein was used, together with an immobilized β -glucosidase, to hydrolyze microcrystalline cellulose (Avicel) to glucose. At the completion of hydrolysis, it was demonstrated that soluble protein could be reabsorbed onto the support and reused for further cellulose hydrolysis.

Microbial Solubilization of Coal

Research has continued on the use of microorganisms to solubilize low-ranked coal. Seven fungi and two bacteria have now been shown to solubilize lignite or treated subbituminous coals (Table 5.4).^{1,2} These are aerobic processes that have primarily been carried out by surface cultures. The bacteria *Streptomyces viridosporus* T7A and *S. setonii* 75Vi2 have more rapid reaction rates than the fungi in both surface culture and suspension

culture (Table 5.5). These biological interactions are apparently catalyzed by an extracellular

Table 5.5. Solubilization of coal by a submerged culture of *S. setonii* 75Vi2

Conditions	Coal ^a	Coal dry weight (mg)		Weight change (%)
		Initial	Final ^b	
Sterile medium	ND	542	517	-4.6
	Wyodak-PT	509	518	+2.0
Submerged cultures ^c	ND	508	289	-43.0
		511	371	-27.5
	Wyodak-PT	513	98	-81.0

^aND is North Dakota lignite; Wyodak-PT is a subbituminous coal pretreated by soaking for 48 h in 8 N HNO₃.

^bFinal dry weights of residual coal after 48-h incubation with cultures.

^cCoal was added to 7-d shake cultures (Sabouraud Maltose medium, 30°C, 100 rpm, 2-in. stroke).

1. C. D. Scott, G. W. Strandberg, and S. N. Lewis, *Biotechnol. Prog.* 2, 131 (1986).

2. G. W. Strandberg and S. N. Lewis, *Biotechnol. Bioeng. Symp.* 17, 153 (1986).

Table 5.4. Microbial species that form a liquid product with low-ranked coal

Microorganism	Source
Fungi	
<i>Trametes versicolor</i> ATCC 12679	Obtained from ATCC
<i>Poria placenta</i> (monticola) ATCC 11538	Obtained from ATCC
<i>Penicillium waksmanii</i> M1.20	Obtained from H. B. Ward, who isolated these organism from Mississippi lignites ^a
<i>Candida</i> sp. ML13	Obtained from H. B. Ward, who isolated these organisms from Mississippi lignites ^a
<i>Aspergillus</i> sp.	Isolated from an as-received lignite sample
<i>Paecilomyces</i> sp.	Isolated from an as-received lignite sample
<i>Sporothrix</i> sp.	Isolated from an as-received lignite sample
Bacteria	
<i>Streptomyces setonii</i> 75Vi2	Obtained from D. L. Crawford, University of Idaho
<i>Streptomyces viridosporus</i> T7A	Obtained from D. L. Crawford, University of Idaho

^aSource: H. B. Ward, *Syst. Appl. Microbiol.* 6, 236 (1985).

component. In the case of *S. setonii*, the extracellular product recovered from culture broths can be used to solubilize coal in the absence of the organism. The bacterium or the extracted microbial product can be effectively utilized for coal solubilization in a shake flask (Table 5.6) or in a fluidized-bed bioreactor (FBR).

Only one coal sample tested, a North Dakota lignite (American Colloid Company, Skokie, IL), was susceptible to rapid and extensive solubilization as received. However, preoxidation of the other coals with a chemical oxidizing agent has been shown to significantly enhance their susceptibility to solubilization. Pretreatment with 8 *N* HNO₃ for 48 to 72 h was found to be the most effective and, therefore, has been used routinely (Table 5.7).²

When analyzed by cross-polarization, magnetic-angle-spinning, dipolar-coupling mass spectrometry [solid state ¹³C nuclear magnetic resonance (NMR)], the lyophilized product from *Candida* sp. interaction with North Dakota lignite exhibited a spectrum qualitatively similar to that of the lignite substrate. The product apparently has a high degree of aromaticity, with an increased number of carboxyl resonances and acid carbonyl or nitrogen groups but a loss of resonance due to methylene groups. There is also an indication of an increase in tetrahedral carbons attached to oxygen. The spectrum of the liquid product is consistent with the hypothesis that the product consists of a mixture of water-soluble hydroxy acids and solubilized coal from which the methylene component has

been removed. Future studies will be directed at the solid state NMR analysis of fractions of the liquid produced by controlled chemical degradations.

5.2 BIOENGINEERING RESEARCH

There has been a continuation of work on the immobilization of microorganisms into biocatalyst beads and the study of columnar bioreactors.

Immobilization of Biocatalysts

The forced flow of alginate or carrageenan solutions through a small nozzle with imposed vibration has been shown to produce monodisperse biocatalyst beads that can be used to entrap microorganisms or enzymes.³ The biocatalyst in this form can be effectively used in fixed-bed or fluidized-bed bioreactors. Since mass transfer of substrates and products within the gel matrix can affect the overall kinetics of the biological process, these effects must be measured and correlated to allow prediction and scaleup.

A technique was developed to measure diffusion coefficients of various materials within the gel beads. The beads were placed into a well-mixed and temperature-controlled vessel in which the decrease (diffusion into the gel) or increase (diffusion out of the gel) of various solutes was measured as a function of time (Fig. 5.6). A

3. C. D. Scott, *Ann. N.Y. Acad. Sci.*, in press.

Table 5.6. The solubilization of coal by cell-free broth from *S. setonii* 75V12

Conditions	Coal ^a	Weight change ^b (%)	Total soluble carbon (mg/L)	Change (mg/L)
Sterile medium	None		20,500	
	ND II	-6.9	20,125	-375
	Wyodak-PT	+2.0	20,375	-125
Cell-free growth broth ^c	None		18,437	
	ND II	-23.0	18,867	+430
	Wyodak-PT	-68.5	20,250	+1813

^aSee Table 5.5 for description of coal samples.

^bWeight of residual coal determined after 48-h incubation (30°C, 100 rpm, 2-in. stroke).

^cBroth from 7-d culture was filter-sterilized using a 0.22-μm Millipore filter.

Table 5.7. Enhancement of fungal liquefaction of coals by preoxidation with nitric acid

Coal	Untreated control	Liquefaction (%) ^a					
		Exposure time (h) ^b					
		HNO ₃ (M)					
		24		48		72	
		5	8	5	8	5	8
Mississippi lignite	<1	38	49	37	66	28	39
Texas lignite	<1	23	28	35	87	69	52
Vermont lignite	<1	79	48	41	37	58	55
Wyodak subbituminous	<1	23	38	24	34	71	73

^aSee Table 5.6 for liquefaction test conditions.

^bExposure to nitric acid at ambient temperature.

Fig. 5.6. Temperature-controlled vessel with well-mixed contents.

computer program was developed to determine the diffusion coefficients by a parameter search technique utilizing the following relationship:

$$\frac{C}{C_0} = \frac{\alpha}{1 + \alpha} + 5\alpha \sum_{n=1}^{\infty} \frac{\exp(-DtQ_n^2/a^2)}{9 + 9\alpha + \alpha^2 Q_n^2}$$

where

- C_0 = initial fluid-phase concentration of solute,
- α = ratio of liquid volume to bead volume,
- Q_n = roots of $(\tan Q_n = 3Q_n/3 + \alpha Q_n^2)$,
- a = bead diameter,
- t = time of contact,
- D = diffusion coefficient.

Diffusion coefficients of low-molecular-weight solutes such as glucose in carrageenan gels without

included biocatalyst approached those in water, while the coefficients in alginate were somewhat less. Higher-molecular-weight substances had lower diffusion coefficients in the gel than in water. The inclusion of microorganisms or other materials in the gel matrix significantly decreased the rate of transfer as shown in Table 5.8.

Advanced Bioreactor Systems

Ethanol Production in a Fluidized-Bed Bioreactor (FBR). The production of ethanol from dextrose by immobilized cells of *Zymomonas mobilis* is being explored in an FBR. This project will attempt to demonstrate the feasibility of this concept and its potential advantages over fed-batch fermentation with yeast. Some optimization of the operating parameters will also be investigated.

This new concept differs from the conventional fed-batch fermentation in three main areas. First, the bacterium *Z. mobilis*, an aerotolerant anaerobe with no minimum oxygen requirement for growth, is substituted for *Saccharomyces cerevisiae*. It can grow somewhat faster and is more ethanol tolerant than the yeast. Second, the organisms are immobilized within gel beads to produce a high cell loading and a correspondingly high biocatalyst activity. The immobilized biocatalysts are retained within the reactor, allowing high conversion at high flow rates. Third, the reactor is operated continuously at flow rates sufficient to fluidize the solid biocatalyst and thus provide high shear force. This improves interphase mass transfer and allows much better disengagement of the CO₂ product gas. In addition, the multistage character of an FBR localizes the effects of product inhibition to the exit region

Table 5.8. Diffusion of glucose in biocatalyst beads at 30°C

Gel material	Solute	Bead diam (cm)	Diffusion coefficient (cm ² /s)
Water	Glucose	NA	6.8×10^{-6}
1% alginate	Glucose	0.33	4.6×10^{-6}
2% alginate	Glucose	0.32	4.9×10^{-6}
3% alginate	Glucose	0.31	4.1×10^{-6}
4% carrageenan	Glucose	0.17	6.3×10^{-6}
4% carrageenan w/3% Fe ₂ O ₃	Glucose	0.10	4.4×10^{-6}
1% alginate w/3% Fe ₂ O ₃	Glucose	0.09	3.8×10^{-6}
4% carrageenan w/ 5.4×10^9 cells/mL	Glucose	0.10	4.1×10^{-6}
4% carrageenan	Albumin	0.10	0.45×10^{-6}
4% carrageenan	Hemoglobin	0.17	0.37×10^{-6}

of the reactor, allowing higher overall reaction rates.

In order to fluidize the beads at practical flow rates, the biocatalyst bead must be relatively small (≤ 2 mm diam). As previously demonstrated, small beads (0.5 to 2 mm) can be produced easily by imposing a harmonic oscillation on a jet of liquid gel which contains cells. The gel droplet is then stabilized in an appropriate salt solution (e.g., K⁺ for κ -carrageenan). From the results above, the diffusion rate does not appear to be an important criterion for selection of the bead composition; therefore, carrageenan was used throughout. Typically, beads of 4% κ -carrageenan and 3% Fe₂O₃ were loaded with concentrated viable cells. The iron oxide was found to be necessary to increase the bead density and decrease bead washout in the FBR.

The response of *Z. mobilis* to typical, unrefined industrial feedstocks, particularly the supplemental nutrients, was unknown and might be a stumbling block to commercialization. Immobilized *Z. mobilis* has been shown in our laboratory to assimilate and to convert industrial feedstocks as well as the laboratory medium. Experiments in both chemostats and in FBRs in which the feed was switched from the typical synthetic feed of 10% glucose, 5% yeast extract, and 0.1 M KCl to an industrial feed of 10% corn dextrose syrup and 20 vol % light steep water (LSW) demonstrated no change in reactor operation from the substitution. Conversion, production, and yield were identical, as can be seen in Fig. 5.7. The A. E. Staley Company has expressed significant interest in the process and

has supplied the industrial feedstocks for this and succeeding experiments from their local ethanol production facility.

A series of tests was made to determine conversion, productivity, longevity, and operability in FBR systems as a function of operating conditions such as flow rate and substrate concentration. Two FBR reactors were used: (1) a small FBR, tapered from 1.25- to 3.81-cm ID in a 122-cm length, that contained 300 mL of biocatalyst beads and (2) a larger column with a 3.81-cm ID that was 225 cm tall and contained 1.4 L of biocatalyst in a 2.3-L reactor. A stabilizing cation (K⁺ for carrageenan) was added as KCl in the feed to maintain a >0.05 M K⁺ concentration. The temperature was maintained at 30°C by a water bath and jacketed columns, and the pH was controlled at 5.0 by the addition of base (KOH) at multiple ports. The overall CO₂ production rate was measured by a wet-test meter, and liquid samples were withdrawn at various ports up the column, filtered, frozen, and then analyzed for glucose and ethanol.

One 30-d test was made in the smaller FBR in which both laboratory and industrial glucose concentrations of 10, 15, and 20% were used at different flow rates, varying from almost packed-bed conditions to maximum productivity. The FBR was operated at each set of operating conditions for about a day.

The overall conversion and productivity data for this series of tests are presented in Fig. 5.8. The highest ethanol concentration was 8.6% at a flow rate of 0.55 L/h and a feed of 17% dextrose. As the reactor was being operated with unsterilized

Fig. 5.7. Glucose concentration in a constant stirred-tank reactor with immobilized *Z. mobilis*. The feed was 10% glucose at $D = 0.5/h$.

Fig. 5.8. Dextrose conversion and ethanol productivity in a medium, laboratory-scale FBR.

feed, the effluent broth contained <0.3 g biomass (dry wt)/L with a small amount of contaminating yeast. The biocatalyst bead surface contained even less yeast, and the washed beads contained internally only *Z. mobilis* at >30 g biomass (dry wt)/L. The biocatalyst can maintain a microaseptic operation even under nonsterile conditions.

An additional advantage of *Z. mobilis* fermentation over yeast is that more of the dextrose is converted to ethanol. The theoretical stoichiometric ratio is 2 mol of ethanol per mol of dextrose. Yeast typically achieves 92% of this ratio, while *Z. mobilis* yields have been reported as high as 99% of the theoretical yield. An average yield of 0.49 g/g or 96% was achieved under a variety of flow rates, feeds, and transients throughout the FBR. This improvement in yield could mean a 5% increase in gallons of ethanol per bushel of corn.

For this new processing approach to be economical, it will be necessary for the biocatalyst beads to have a reasonable lifetime before replacement is required. The longevity of biocatalyst beads was studied in FBRs. Beads were continuously fluidized with unsterilized feed for 2 months and remained actively fermenting and stable throughout this period. Preliminary cost estimates indicate that with an active biocatalyst life of >60 d, the biocatalyst cost becomes a negligible portion of the ethanol cost.

In the larger reactor, experiments were performed as before to determine the productivity and conversion as a function of flow rate and feed concentration. In both reactors, it is clear that, of the operating conditions used, a feed with 10% dextrose consistently gives the highest conversion rates; however, a feed with 15% dextrose can also be used for equally high conversions at lower flow rates. The highest productivities and ethanol concentrations were obtained with 15% glucose feed. A feed with 20% dextrose resulted in decreases in both conversion and productivity, which is consistent with earlier tests made with free cells where it was observed that substrate inhibition becomes important at a glucose concentration of 16%. Selected results, listed in Table 5.9, show that enhanced productivities and high conversions can be achieved at the same time in the immobilized-cell FBR. Typical axial concentration profiles of glucose and ethanol from the larger FBR are presented in Fig. 5.9, illustrating the multistage character of an FBR.

The immobilized-cell FBR achieved enhanced productivities of 50 to 100 g EtOH/L·h during a

Table 5.9. Selected data from a large laboratory-scale FBR with Staley feed

Dextrose concentration		Feed rate (L/h)	Productivity (g EtOH/L·h)	Conversion (%)
Feed (g/L)	Effluent (g/L)			
101	0.6	2.12	48.8	99.4
96*	0.0	2.01	47.5	100.0
153	1.2	2.28	75.2	99.2
143*	0.4	1.06	25.6	99.7

*Data points not included in Fig. 5.8 due to deviation from desired feed %.

Fig. 5.9. Axial concentration profile in an FBR.

fourfold increase of scale over previous laboratory tests. These productivities can be accomplished with effluent residual dextrose concentrations of $<0.1\%$. At 15% dextrose feed, $>7\%$ ethanol is achievable. These results indicate that an effluent stream comparable to batch fermentation is achievable at a more than tenfold increase in productivity. Long-term activity of the biocatalyst has been demonstrated to be at least 2 months.

These results are encouraging for the utility of the process and for further exploration of the optimal conditions and scaleup.

Mass Transfer Effects. In general, diffusional resistances within biocatalyst beads will tend to reduce the transport rates of substrates and products and thus affect the overall biocatalyst kinetics. In most cases, this will lead to a decrease of reaction rate unless very small beads are used. However, if there is significant substrate inhibition of the biological kinetics, the mass transport resistance of the catalyst may result in a sufficiently low concentration of the substrate to give an

apparent enhancement in the innate kinetics. Both theoretical and experimental studies of this phenomenon are being carried out.

Typically, the problem of diffusion coupled with a chemical reaction is solved by iterative integration of the two-point, boundary-value problem. An alternative approach is to approximate the concentration profile inside the biocatalyst bead as a summation of orthogonal polynomials. Orthogonal collocation reduces the problem to a set of algebraic equations, by which the effectiveness factor of the catalyst, η , can be estimated as a weighted sum of the reaction rate at the collocation points,

where

$$\eta = \frac{\text{Overall reaction rate}}{\text{Reaction rate at bulk conditions}} \quad (2)$$

A critical parameter is the Thiele modulus, defined as

$$\phi^2 = \frac{(Q_{p_m}/Y_{ps}) R^2 X}{D_s S_0} \quad (3)$$

where

- Q_{p_m} = the maximum specific ethanol production rate,
- Y_{ps} = the yield of EtOH/g glucose,
- R = radius,
- X = biomass,
- D_s = diffusion coefficient of glucose,
- S_0 = the bulk substrate.

For $\phi^2 < 10$, three collocation points are sufficient. Calculations showed that at a Thiele modulus of ~ 7 , there is an appreciable enhancement of the overall reaction rate at high S_0 values due to the consumption of the inhibiting substrate within the bead. That is, the substrate is consumed as it is transported through the bead, thus reducing the inhibition and resulting in an increase in the reaction rate relative to the bulk substrate concentration. Eventually, in large beads ($\phi^2 > 10$), the resistance to mass transport results in a depletion substrate at the bead center and the effectiveness factor falls again.

Experiments were performed with a small number of biocatalyst beads in a differential fluidized reactor in which all of the biocatalyst can be assumed to be at inlet concentration. Beads ranging in diameter from 1 to 5 mm were used in

separate experiments at glucose concentrations of 10, 15, and 20%. At least 24 h was allowed for microbial adaptation when the concentration was changed.

The specific production formation rate, Q_p , was calculated from the experiments and was found to be similar to those determined on a theoretical basis (Table 5.10). They show that as the Thiele modulus is increased, the effectiveness factor also increases until a maximum occurs at $\phi^2 \sim 6$. This confirms the use of the Thiele modulus to characterize biocatalyst systems. The amount of enhancement is somewhat greater than predicted. In addition, it can be observed in Table 5.10 that a change in flow rate resulted in a negligible change in the effectiveness factor. This is convincing evidence that external mass transfer resistances are negligible under fluidization.

Table 5.10. Effectiveness factors under substrate inhibition for 4-mm beads immobilizing *Z. mobilis* at a concentration of 15 g (dry wt)/L.

Substrate (g/L)	Flow rate (L/h)	Bead diam (mm)	ϕ^2	η
200	0.51	1.57	0.93	1.02
150	0.51	1.57	1.06	1.03
100	0.51	1.57	1.36	1.14
200	0.64	4.10	3.68	1.14
150	0.64	4.10	4.40	1.21
150	1.18	4.10	4.40	1.19
100	0.64	4.10	6.00	1.45
100	1.16	4.10	6.05	1.45
100	0.64	4.10	8.06	1.12
100	1.16	4.10	8.16	1.09

While larger beads are seen to enhance apparent reaction rates slightly in substrate-inhibited regimes, this advantage will be outweighed by the high Thiele modulus at low-substrate and high-product regimes that occur in other parts of a bioreactor system. Thus, small beads are still recommended when high conversion is desired. In this light, a beneficial sidelight of fluidization is its natural sorting action of bead sizes, which will congregate the largest beads at the bottom of the column where the substrate concentration is the highest. This might suggest use of multiple bead sizes in an integrated reactor system.

FBR Hydrodynamics. In order to design an FBR, it will be necessary to predict the kinetics within the biocatalyst particles and the mass transport to and within the beads. In addition, there

must be knowledge of the distribution of the fluidized biocatalyst as well as of the gas and liquid phases within the reactor.

Electroconductivity is being explored as a technique to measure the gas holdup with this three-phase FBR system. In essence, both the broth and the beads are conductive, due to the salts present, while the gas is not. Therefore, the conductivity will decrease linearly with the amount of gas present. The effects of glucose, ethanol, and cell concentration on the conductivity of a 0.1 M KCl solution at 30°C were measured to be linear for each and <20% in the range of interest. This allowed a correlation to be written for the liquid conductivity as a function of other measurable system variables.

The conductivity of the 4% κ -carrageenan beads, which was measured in a 0.1 M KCl solution at 30°C in a 1-in.-diam column, was 80% of the bulk solution. In an actively fermenting FBR, the conductivity meter did respond directly to passing bubbles, allowing a rough estimate of the gas phase by neglecting the difference in the liquid and bead conductivity.

The hydrodynamics of a gas-producing, three-phase fluidized bed caused transient variations in measurements in laboratory-scale columns. In a small, fermenting fluidized bed, the produced gas (CO₂) forms clumps or fronts of bubbles that move through the bed together without slugging the solids. These fronts are directly associated with oscillations of up to $\pm 5\%$ in the substrate and product concentrations within the column. While undetectable at the top, these fluctuations are large and measurable midway in the column. It is believed that these phenomena will become less important in larger columns.

5.3 BIOANALYTICAL RESEARCH

The objective of this program is to develop instrumentation and methodology for measuring toxic compounds in the environment and for measuring the exposure of personnel to toxic materials. A portable fluorometric monitor was developed earlier in this program to measure polynuclear aromatic compounds on inanimate surfaces and on skin.⁴ A microfluorometric technique was also designed to measure mixed-function oxidase (MFO) activity in various organs in mice exposed to toxic materials.⁵⁻⁷ These measurements have been expanded to determine the induction of

MFO activity in mouse lymphoid tissues and in bluegill sunfish

Because of the inherent difficulty of studying a heterogeneous human population, we investigated MFO activity measurements as a biomarker of polycyclic aromatic hydrocarbon (PAH) exposure in lymphoid tissues in an inbred mouse model system. This study⁸ attempted to determine (1) whether systemic administration of PAHs can induce MFO activity in lymphoid tissues and (2) which of the lymphoid tissues might be so induced. The mice used in these studies were 8- to 16-week-old females of the C3H/HeNCrLBR strain. Benzo[a]pyrene (BaP) and 3-methylcholanthrene (3-MC) were dissolved in corn oil and administered by intraperitoneal (ip) injection. Control animals received ip injections of an equal volume by corn oil. Animals were sacrificed by cervical dislocation; spleen, thymus, liver, and lung were removed and processed for analysis.

The MFO activities in 'S-9' fractions of mouse spleen after various PAH treatments are shown in Table 5.11. The mean activity values averaged from individual animals are reported along with the standard deviation of the mean. The MFO activity measurements on 'S-9' samples from individual spleens were made in triplicate. The enzyme activity data in Table 5.11 indicate that, of the times after PAH treatment at which MFO activity was measured, significant MFO induction occurred only at 48 h. In the case of either BaP or 3-MC, 16 h following PAH treatment was insufficient time to produce a significant increase in MFO activity. We have reported MFO activities for two separate 48-h BaP data sets. Activity for the first data set is reported as picomoles per minute per

4. C. A. Burtis and J. E. Mrochek, *Development of a Portable Fluorescence Monitor to Detect Surface Contamination by Polycyclic Hydrocarbons*, ORNL/TM-8753, November 1983.

5. *Chemical Technology Division Progress Report for the Period April 1, 1983, to March 31, 1985*, ORNL-6153, October 1985.

6. B. Z. Egan, N. E. Lee, C. A. Burtis, J. Y. Kao, and J. M. Holland, "Use of Laser-Excited Fluorescence to Measure Mixed-Function Oxidase Activity," *Clin. Chem.* **29**, 1616 (1983).

7. C. A. Burtis, B. Z. Egan, and N. E. Lee, "Development of Laser-Excited Fluorometric Assays for the Monitoring of Toxic Chemical Exposure," *Pure Appl. Chem.* **57**, 543 (1985).

8. G. D. Griffin, B. Z. Egan, N. E. Lee, and C. A. Burtis, "Induction of Mixed-Function Oxidase Activity in Mouse Lymphoid Tissues by Polycyclic Aromatic Hydrocarbons," *J. Toxicol. Environ. Health* **19**, 185 (1986).

Table 5.11. MFO activity^a in C3H mouse spleen 'S-9' fractions following intraperitoneal PAH administration

Treatment	Time after PAH treatment when MFO activity was assayed					
	16 h		48 h ^c		48 h ^c	
	Activity ^b	Ratio (I/C) ^d	Activity	Ratio (I/C)	Activity	Ratio (I/C)
Control	0.28 ± 0.12 (4)		0.18 ± 0.06 (5)		12 ± 4 (5)	
Benzo(a)pyrene ^e	0.44 ± 0.29 (5)	1.6	0.89 ± 0.13 (3) ^g	4.9	45 ± 15 (8) ^h	3.8
3-Methylcholanthrene ^e	0.27 ± 0.03 (3)	0.96	0.45 ± 0.11 (3) ^f	2.5		

^aMean ± SD; number of animals in parentheses.

^bAs pmol resorufin formed/min·mg 'S-9' protein.

^cRatio of the PAH-induced/control MFO activity.

^dIn this set of experiments, the activities are expressed as pmol product formed/min·mg 'S-9' protein.

^eIn this set of experiments, the activity values are shown for pmol product formed/min·g tissue.

^fBaP dose = 180 mg/kg body weight (4.6 mg/mouse).

^gp = 0.0002.

^hp = 0.0001.

ⁱ3-MC dose = 160 mg/kg body weight. (4 mg/mouse).

^jp = 0.0016.

milligram protein; for the second data set, it is reported as picomoles per minute per gram tissue (based on original weight of spleen). By either activity measurement, the MFO activity in spleen was induced 48 h after ip BaP administration. The same conclusion may be drawn from the results obtained when 3-MC was used as the inducer. Although the induction ratio with 3-MC is not as large as that with BaP for the 48-h activity measurement (2.5 vs 4.9), no definitive conclusions about the relative efficacy of BaP or 3-MC as an MFO inducer can be drawn from the limited data. A more comprehensive study of MFO induction in response to PAH treatment would be necessary to determine the exact time at which enzyme induction is maximal.

Enzyme activity measurements were performed on selected tissues to determine whether MFO induction occurs in other lymphoid tissues following systemic PAH treatment. Due to the small amount of thymus tissue available from an individual animal and the difficulty of homogenizing this small amount of tissue, thymus homogenates were prepared only in experiments in which a "pooled" thymus sample from a reasonable number of animals could be collected (e.g., the composite experiment). The total weight of thymus tissue from the five control animals in the composite experiment was 0.121 g, and the thymus weight for the 16 PAH-treated animals was 0.408 g. No MFO activity could be detected in either the microsomes or the 'S-9' fraction from the control

thymus preparation. Activity was found, however, in both microsomal and 'S-9' fractions from 3-MC-induced animals (1.4 pmol/min·mg for microsomes and 0.28 pmol/min·mg for the 'S-9' fraction). In another experiment, a pooled thymus sample (0.105 g) was prepared from five animals 48 h following BaP treatment. The MFO activity of the 'S-9' fraction from this sample, 43 pmol product/min·g tissue, was quite similar to that for the spleen 'S-9' from the same group of animals. These results indicate that mouse lymphoid tissue cells can respond to chemical insults by an MFO induction response.

The MFO activity can also be induced in fish exposed to xenobiotic compounds, such as BaP, in aquatic environments. In collaboration with personnel of the ORNL Environmental Sciences Division, we have measured the MFO activity in fish liver samples obtained from fish in various environments. Variables such as temperature, diet, and BaP concentration affect the MFO activity in the liver of the fish. Fish that were denied food and acclimated to 4, 13, and 26°C for 2 weeks exhibited low, relatively uniform MFO activity. However, fish that were fed and acclimated to 26°C showed 7 times higher MFO activity than those acclimated to 4°C. When starved fish were acclimated at 26°C and injected with BaP (20 mg BaP per kilogram of body weight), the liver MFO activity increased by a factor of 30. The MFO activity level was also affected by the type of food in the diet. The highest MFO induction occurred

on a red worm diet, with trout chow yielding an intermediate MFO induction and crickets producing no MFO induction. In the pH range of 5 to 9, no significant effect of water pH on the MFO activity in fish occurred. These results suggest that

environmental variables such as nutritional status and season should be taken into account if MFO activity in fish is to be used as a biological indicator of pollution.

6. TRANSURANIUM-ELEMENT PROCESSING

The Transuranium Processing Plant (TPP) is the production, storage, and distribution center for materials used in the heavy-element research program in the United States. The isotopes produced at the TPP continue to be used nationally and internationally in basic studies of the physics and chemistry of the transuranium elements, including experiments designed to produce new, neutron-rich isotopes of the heaviest elements. The TPP also collaborates with other divisions at ORNL and with other national laboratories in research on actinide and transactinide elements. In addition, the TPP is the sole source of the ^{252}Cf that is distributed worldwide by the DOE's ^{252}Cf Industrial Sales/Loan Program, which was recently transferred from the Savannah River Laboratory to ORNL. The ^{252}Cf is used to prepare neutron sources for a wide variety of applications in both industry and medicine.

6.1 TPP OPERATIONS

Target rods fabricated at the TPP and irradiated in the High Flux Isotope Reactor (HFIR) are pro-

cessed at the TPP for separation, recovery, and purification of the heavy actinide elements, which are distributed to laboratories throughout the country for use in research work. Products are usually highly purified prior to shipment and are frequently provided in special chemical forms and/or in special devices required by the experimenter.

Production and Shipment

During this report period, 12 HFIR targets were processed to recover the transplutonium elements. Table 6.1 presents data on the amounts of key isotopes obtained during this 21-month period, as well as the total production of each isotope at the TPP in 21 years of operation.

Fifty-three shipments of transuranium elements were made during the report period. The total quantities of each type of product shipped are listed in Table 6.2.

Flowsheets and process equipment used at the TPP remained unchanged during this report period.

Table 6.1. Materials recovered in the Transuranium Processing Plant

	April 1985 through December 1986	Total through December 1986
Number of years	2	21
Number of target campaigns	1	47
Number of targets processed	12	552 ^a
Amounts of key isotopes recovered:		
^{242}Pu	0	141 g
^{243}Am	2.3 g	294 g
^{244}Cm	61 (106) ^b g	2302 g
^{249}Bk	36 mg	768 mg
^{252}Cf	416 mg	7262 mg
^{253}Es	2.3 mg	32 mg
^{257}Fm	~2 pg	18 pg ^c

^aThe number of targets includes 355 HFIR targets, 176 SRP slugs and targets, and 21 SRP Pu-Al tubes.

^bThe value in parentheses is the total mass of all curium isotopes.

^cEstimated amount.

Table 6.2. Transuranium elements shipped during the period April 1, 1985 through December 31, 1986

Major isotope	Total quantity	Number of shipments
^{240}Pu (99%)	13.5 g	1
^{241}Am	0.3 g	1
^{242}Cm (99%)	25.5 μg	1
^{244}Cm (91%)	29 g	3
^{246}Cm (97%)	113 mg	3
^{249}Bk	36 mg	6
^{250}Cf	19.6 mg	5
^{252}Cf	0.6 mg	1
^{252}Cf	356 mg	20
^{253}Es (mixed)	74 μg	3
^{253}Es (milked)	173 μg	1
^{254}Es	8.3 μg	3
^{255}Fm	7.6 ng	4
^{257}Fm	1.9 pg	1

Target Fabrication

Twelve HFIR targets for transuranium-element production were fabricated from a mixture of curium and americium oxides. Feed materials for these targets included recycled americium-curium mixtures recovered from irradiated HFIR targets and ~ 47 g of curium that had originally been produced in the Savannah River Curium II Program for DOE's Space Nuclear Systems Program.

In addition, four special targets were made in collaboration with ORNL's Operations Division; these targets included three containing ^{62}Ni and one containing both ^{54}Fe and ^{118}Sn .

6.2 LARGE EINSTEINIUM ACTIVATION PROGRAM (LEAP)

Four national laboratories active in transplutonium research [Lawrence Berkeley Laboratory (LBL), Los Alamos National Laboratory (LANL), Lawrence Livermore National Laboratory (LLNL), and ORNL] have collaborated to propose a major new thrust, the Large Einsteinium Activation Program (LEAP). This will require the production of about ten times as much ^{254}Es as the 3 to 4 μg that is currently available. A sample of ~ 40 μg would open new opportunities for research on the almost unknown chemical properties of elements 102 through 105 and the nuclear properties of nuclides having atomic numbers at least as high as 109. It may even be possible to approach the calculated "island of stability" for superheavy elements.

This new initiative is critically dependent upon the capability of the HFIR/TPP complex to produce ~ 40 μg of ^{254}Es . Production of this amount of material in the HFIR by normal irradiation of standard TPP/HFIR targets is impractical because of the very low yields of such a highly fissionable isotope.

The cross-section data indicate that reactions in the epithermal region are much more favorable than reactions in the thermal energy region for ^{254}Es production. The key to enhancing the production of ^{254}Es in the HFIR is the use of cadmium filters¹ around ^{252}Cf targets to diminish the thermal component of the neutron spectrum.

An irradiation scheme has been developed which takes into account the HFIR operating schedule, the time scale of the transmutation reactions, and the use of cadmium filters. This irradiation scheme is depicted in Fig. 6.1. Targets containing ^{252}Cf are irradiated in the large beryllium reflector region of the HFIR for one normal fuel assembly lifetime, ~ 21 d, to build up an inventory of ^{253}Cf . The targets are removed from the reactor for ~ 20 d to allow ^{253}Es to be formed by decay of the ^{253}Cf ; the targets are then irradiated for ~ 5 d in the target island at maximum thermal flux to restore the inventory of ^{253}Cf . This establishes a suitable ratio of ^{253}Cf and ^{253}Es for the final stage of irradiation. The targets are then placed in cadmium filters and irradiated 24 to 28 d in the HFIR target island to convert ^{253}Es to ^{254}Es by neutron capture. The cadmium filters screen the target from the thermal flux and eliminate the consumption of ^{254}Es through thermal fission. Several reactor shutdowns are required during the final stage to replace the cadmium filters and spent fuel assemblies.

The presence of cadmium in the HFIR target island introduces several technical considerations. The high thermal-neutron cross section of ^{113}Cd that makes it a good filter will also cause it to be readily consumed in the high thermal flux of the HFIR. The cadmium used to surround the HFIR targets must be thick enough to have a useful lifetime, but the total amount of cadmium in the target island must be kept small enough to permit the reactor core to achieve and maintain criticality for

1. J. E. Bigelow, C. W. Alexander, and L. J. King, "Proposed Production of a Large (~ 40 - μg) Sample of ^{254}Es ," presented at the Symposium on Heavy Element Research, AIChE 1985 Summer National Meeting, Seattle, Washington, August 25-28, 1985.

Fig. 6.1. Target composition changes during irradiation of ^{252}Cf to produce a large ^{254}Es sample.

a useful fraction of the normal fuel lifetime. The cadmium will also cause a thermal flux depression in the target island, and the core's power distribution will be shifted.

With the assistance of ORNL's Computing and Telecommunications Division, one- and three-dimensional reactor calculations are being made to examine these effects. To date, calculations indicate that there is sufficient cadmium to overcome the burnup effects, and the negative reactivity of the ^{113}Cd is not as severe as originally expected. Because the negative reactivity is relatively small, there is little control rod movement, resulting in only a moderate change in the HFIR core's power distribution. Flux depression effects in the target island are still being examined.

The burnup of ^{113}Cd is also being examined experimentally. Four cadmium cylinders were irradiated in the HFIR's hydraulic tube with various flux and time parameters. Cobalt flux wires inside the cylinders seemed to show only an epithermal neutron response, indicating that total burnout of the ^{113}Cd had not occurred. In an attempt to determine the extent of the burnup of ^{113}Cd as a function of the various parameters, thin layers (50–100 μm) of the cadmium cylinders will be removed and

analyzed for the 113/110 mass ratio. This should ultimately result in a correlation between flux and the effective lifetime of ^{113}Cd in HFIR. A short-term, full-scale, irradiation test is planned to validate the calculational results¹ indicating that an ^{254}Es sample of suitable size can be produced using the cadmium filters. Various activities have begun in preparation for the irradiation test.

One of the major concerns is the ability of HFIR to remove radiation-generated heat from the LEAP targets. In preparation for heat-removal testing, a prototype LEAP assembly (see Fig. 6.2) was fabricated and subjected to hydraulic conditions identical to those in the HFIR target island. The assembly met design requirements and was hydraulically qualified for use in HFIR. Three additional LEAP assemblies, including trefoils and cadmium filters, have been fabricated for use in the irradiation test. While there are some remaining fabrication difficulties with the cadmium filters, these are not expected to interfere with the irradiation test.

Dummy target pellets, which will hold flux wires, have also been fabricated. These will be inserted into the LEAP assemblies and/or other dummy HFIR targets in order to map flux depres-

Fig. 6.2. Components of the HFIR LEAP target assembly.

sion in the target island during the test. Some computer codes have been written to determine the two-group neutron fluxes and associated errors from the various flux wire responses.

Remote handling equipment to be used for underwater transfers of the LEAP assemblies has been designed and fabricated. Cold testing of this equipment is pending.

Work that remains to be completed prior to the irradiation test includes the fabrication of test pellets and target rods and a review of the proposed experiment.

6.3 SPECIAL PROJECTS

The facilities at the TPP are used for a variety of purposes in addition to those associated with the mainline production and distribution of transuranium elements. These special projects include nonroutine isotope productions, unusual nuclide preparations, special irradiations in the HFIR, and the fabrication of neutron sources from ^{252}Cf .

Fabrication of Neutron Sources from ^{252}Cf

Some of the californium recovered at the TPP is incorporated into neutron sources that are loaned to researchers. Three of these sources, containing from 7 to 25 mg of ^{252}Cf , were fabricated during this report period. All were doubly encapsulated in

type 304L stainless steel in a standard TPP configuration.²

Allocation of Neutron Sources

Neutron sources are returned to the TPP when the projects for which they were requested are completed or when replacements are ordered following decay of the ^{252}Cf . These sources are available for reassignment until the timing is appropriate for reprocessing to recover the ^{248}Cm daughter. During the current report period, 12 sources were returned and 10 were reassigned; 2 were withdrawn from the loan program to make their unusual isotopic content available for research; 82 are currently on loan; and 29 (containing $\sim 4\ \mu\text{g}$ to 5.2 mg of ^{252}Cf) are available for reassignment. Four of the reassigned ^{252}Cf neutron sources, totaling 550 μg , were sent to Brookhaven National Laboratory for use in the development of in vivo neutron activation analysis techniques for clinical diagnosis and for monitoring medical conditions involving abnormal quantities of lithium or aluminum.

2. J. E. Van Cleve, Jr., L. C. Williams, J. B. Knauer, and J. E. Bigelow, "Fabrication of ^{252}Cf Neutron Sources at Oak Ridge National Laboratory," *Applications of Californium-252, Proc. of Amer. Nucl. Soc. Topical Meeting, CONF-720902*, U.S. Energy Research and Development Administration, Washington, D.C., 1972, p. 25.

Electroplated ^{252}Cf Fission Sources

Three ^{252}Cf fission sources were prepared during this report period by electroplating small amounts of purified californium from dry isopropanol solutions onto platinum foils or disks; these sources contained from 11 to 950 mg of ^{252}Cf .

One electroplated source of ^{252}Cf on platinum, containing 11.1 μg , was prepared for the ORNL Isotopes Research Materials Laboratory. Such plates are used to provide self-transfer ("mass-free") fission sources in several geometries for a variety of experiments.

Two electroplated ^{252}Cf sources on platinum, each containing nearly 1 μg , were prepared for EG&G Idaho for use in an ongoing program to discover new short-lived nuclides and measure their decay properties. In these experiments, the spontaneous fission fragments from the californium are ejected from the plates by recoil, trapped on aerosol particles, and carried away from the source plates by a helium stream to an inline mass spectrometer.

Preparation of ^{242}Cm

A batch of ^{242}Cm was prepared by irradiating ~ 5.5 mg of ^{241}Am in the HFIR for 16 h. After a decay period of several days to permit the 16-h $^{242\text{m}}\text{Am}$ to decay completely to ^{242}Cm , the target material was dissolved and processed through a pressurized ion-exchange column and then through a micro ion-exchange separation to produce a 50- μg lot of 99.7% isotopic purity ^{242}Cm , with ≤ 180 ppm residual ^{241}Am .

Approximately half of the ^{242}Cm product was sent to LLNL. There it was used to prepare a small target for the Rensselaer Polytechnic Institute Accelerator, and a most successful experiment was performed to measure the energy-dependent fission cross section.³

Preparation of ^{250}Cf

Hydraulic rabbit capsules containing 3.0 mg of ^{249}Bk were irradiated in the HFIR to produce ~ 0.627 mg of ^{250}Cf , which was then isolated and purified. The ^{250}Cf was sent to LBL for spectrographic studies of isotope shift effects.

The residual ^{249}Bk from this preparation was reirradiated at the request of LBL to produce another ~ 0.5 mg of ^{250}Cf , which was isolated and purified and will be reirradiated during the LEAP irradiation test to produce a sample of ^{251}Cf .

Preparation of ^{255}Fm

Approximately 7.6 ng of high-purity ^{255}Fm has been recovered from the einsteinium product of Campaign 66. Two shipments totaling 4.4 ng were transferred to the Transuranium Research Laboratory of ORNL's Chemistry Division. This material was used to study the inorganic chemistry of fermium. The remaining 3.2 ng of ^{255}Fm was sent in two lots to the Oak Ridge Associated Universities Medical Division for studies of radiolabeled monoclonal antibodies as a possible diagnostic and/or therapeutic tool in the treatment of colo-rectal cancer.

The ^{255}Fm (20-h half-life) is the decay daughter of ^{255}Es (40-d half-life), which is present in small quantities in the einsteinium product from irradiated HFIR targets ($\sim 0.03\%$ of the total einsteinium). After the primary fermium product (which contains ^{257}Fm) was separated from the einsteinium product of Campaign 66, the einsteinium was stored to permit ^{255}Fm to grow in by radioactive decay. The ^{255}Fm was then recovered and purified by a series of pressurized, ion-exchange operations.

3. B. Alam, R. C. Block, R. E. Slovacek, and R. W. Hoff, "Measurement of the Fission Cross Section of ^{242}Cm ," *Trans. Amer. Nucl. Soc.* 53, 467-68 (1986).

7. NUCLEAR REGULATORY COMMISSION PROGRAMS

The U.S. Nuclear Regulatory Commission (NRC) programs in Chem Tech are focused on two diverse tasks: nuclear reactor safety [primarily light-water reactors (LWRs)] and nuclear waste repository licensing. These two activities are discussed in the following sections.

7.1 NUCLEAR REACTOR SAFETY

Current NRC reactor safety programs emphasize research on the mechanisms and pathways by which fission products are released to the environment from failed fuel elements under hypothetical reactor accident conditions. The following sections discuss programs for the investigation of fission product release from LWR fuel, postaccident iodine and tellurium chemistry, the TRAP-MELT code validation and aerosol resuspension, and support for the LWR Aerosol Containment Experiment (LACE).

Fission Product Release from LWR Fuel

The large quantities of radioactive fission products and transuranics in the irradiated fuel comprise the principal hazards associated with nuclear reactors. Accordingly, reliable assessment of the consequences of reactor accidents requires detailed information on the release and transport behavior of these radioactive materials.¹ Such behavioral characteristics depend on temperature, time, and atmosphere (oxidizing or reducing), as well as on the irradiation history of the fuel (burnup and heat rating). Tests of commercial LWR fuel, sponsored by the NRC, have been conducted in a hot cell facility at ORNL for several years.²⁻⁶ In addition to measurement of the fractional release of the more volatile fission products, especially Kr, I, and Cs, the objectives of this work have included studies of the chemical forms of these and other elements released under accident

conditions.^{7,8} Toward this end, the tests of commercial fuel have been supplemented by tests utilizing specific chemical forms of fission product tracers with unirradiated fuel.

Early in this reporting period the experimental equipment was rebuilt, converting the furnace from a horizontal to a vertical orientation (to better simulate the vertical arrangement of LWR fuel rods) and improving temperature measurement and

1. *Technical Bases for Estimating Fission Product Behavior During LWR Accidents*, NUREG-0772, U.S. Nuclear Regulatory Commission, June 1981.

2. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, O. L. Kirkland, and R. L. Towns, *Fission Product Release from Highly Irradiated LWR Fuel*, NUREG/CR-0722 (ORNL/NUREG/TM-287/R2), February 1980.

3. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, M. F. Osborne, and R. L. Towns, *Fission Product Release from Highly Irradiated LWR Fuel Heated to 1300-1600°C in Steam*, NUREG/CR-1386 (ORNL/NUREG/TM-346), November 1980.

4. R. A. Lorenz, J. L. Collins, M. F. Osborne, R. L. Towns, and A. P. Malinauskas, *Fission Product Release from BWR Fuel Under LOCA Conditions*, NUREG/CR-1773 (ORNL/NUREG/TM-388), July 1981.

5. M. F. Osborne, J. L. Collins, and R. A. Lorenz, "Fission Product Release from UO₂ Under LWR Accident Conditions: Recent Data Compared with Review Values," presented at 87th Annual Meeting of American Ceramic Society, May 5-9, 1985, Cincinnati, Ohio; published in *Advances in Ceramics* 17, 191-97 (1986).

6. M. F. Osborne, J. L. Collins, R. A. Lorenz, and R. V. Strain, "Fission Product Release and Fuel Behavior in Tests of LWR Fuel Under Accident Conditions," in *Proceedings of the International Symposium on Source Term Evaluation for Accident Conditions*, Columbus, Ohio, Oct. 28-Nov. 1, 1985, International Atomic Energy Agency, Vienna, Austria, March 1986.

7. J. L. Collins, M. F. Osborne, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, *Observed Behavior of Cesium, Iodine, and Tellurium in the ORNL Fission Product Release Program*, NUREG/CR-3930 (ORNL/TM-9316), February 1985.

8. J. L. Collins, M. F. Osborne, and R. A. Lorenz, "Fission Product Tellurium Release Behavior Under Severe Light Water Reactor Accident Conditions," *Nucl. Technol.* 77(1), 18-31 (1987).

control capability.⁹ The new furnace design, which is capable of operation at 2,700 K, is shown in Fig. 7.1. Additional equipment was installed to allow continuous measurement of the hydrogen generated by the steam-Zircaloy reaction.

Two experiments were conducted in the vertical test apparatus, and the results are being evaluated. Test conditions and preliminary results are summarized in Table 7.1. These limited fission product release data are generally consistent with the results of the previously reported horizontal tests.¹⁰ No significant differences resulting from the change in orientation were apparent. The longer-duration test (VI-2), however, confirmed a significant reduction in release rate with increasing time (fraction of inventory released), which will strongly influence extrapolations of the results from the earlier 20-min tests. Release rate data for ^{137}Cs are shown in Fig. 7.2; the results from tests VI-1 and VI-2, and also from the HI test series, are compared with the generally accepted CORSOR-M

model,¹¹ which was based on the earlier NRC-sponsored review.¹ The open points with connecting arrows in Fig. 7.2 indicate the progression of the ^{137}Cs release rate during the two most recent tests; in both cases the release rate declined relative to the accepted standards.

The results of these experiments have provided valuable information about the release behavior of several of the most important fission products, especially I, Cs, Te, and Sb. For instance, in the

9. M. F. Osborne, J. L. Collins, P. A. Haas, R. A. Lorenz, J. R. Travis, and C. S. Webster, *Design and Final Safety Analysis Report for Vertical Furnace Fission Product Release Apparatus in Hot Cell B, Building 4501*, NUREG/CR-4332 (ORNL/TM-9720), March 1986.

10. *Chemical Technology Division Progress Report for the Period April 1, 1983, to March 31, 1985*, ORNL-6153, October 1985.

11. M. R. Kuhlman, D. J. Lehmickie, and R. O. Meyer, *CORSOR User's Manual*, NUREG/CR-4173 (BMT-2122), Battelle Memorial Institute, March 1985.

Fig. 7.1. Vertical fission product release furnace.

Table 7.1. Test conditions and preliminary results of vertical fission product release tests

Parameter	Test VI-1	Test VI-2
Fuel specimen:^a		
Reactor (pressurized-water reactor)	Oconee	BR3
Burnup, MWd/MTIHM ^b	38,300	40,000
Test temperature(s), K	2,020; 2,300	2,300
Test times, min	20, 20	60
Time at >2,000 K, min	54	70
Gas flow rates, L/min at STP:		
Steam into furnace	1.8	1.4
Helium into furnace	0.7	0.5
Fission product release, %		
¹³⁷ I ^c	62	>30
¹³⁷ I ^d	39	e
¹³⁷ Cs	62	58/
¹²⁵ Sb	12	e

^aBoth fuel specimens were 15.2 cm long.

^bMegawatt-days per metric ton of initial heavy metal.

^cValues are based on the inventory in the fuel specimen as determined by ORIGEN calculation.

^dThis includes gas released during irradiation.

^eData are not yet available.

^fThis value is uncertain because of the difference in the two measurement methods that are still being evaluated; it may be as high as 68% or as low as 48%.

steam-hydrogen environment, iodine behaves as CsI, and cesium behaves as the relatively unreactive CsI and the highly reactive CsOH. Antimony behaves as the elemental vapor, readily depositing on and combining with other metals. Tellurium combines with, and is retained by, the metallic Zircaloy cladding but is released upon complete oxidation of the cladding, apparently as SnTe or possibly as CsTe.

In a second part of the fission product release program, the core-melt test apparatus is used. This apparatus inductively heats, in flowing steam, a 61-rod bundle of unirradiated fuel rods that contains more than 10 kg of fuel, cladding, control rod material, grid spacers, and simulated fission products. Test bundles were heated as high as the melting point of the fuel-cladding-spacer mixture, and the release of fission products and control rod materials was measured. The results were used to predict total releases of fission products from an LWR core for a severe accident.^{12,13} It was found that the release of silver from the control rod alloy was much less than that expected from its vapor pressure because of rundown to cooler locations in the heated bundle (reactor core).

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Fig. 7.2. Comparison of release rates for ¹³⁷Cs from VI and HI tests with the CORSOR-M model and the NUREG-0772 curve.

Postaccident Iodine and Tellurium Chemistry

Chemistry and Transport of Iodine in Containment. The ultimate aim of a description of iodine behavior in severe LWR accidents is a time-dependent accounting of iodine species released into the containment and to the environment. Factors involved in the behavior of iodine can be conveniently divided into four general categories:

12. G. W. Parker, G. E. Creek, and A. L. Sutton, Jr., "Source Term Evaluations from Recent Core-Melt Experiments," in *Proceedings of the International Symposium on Source Term Evaluation for Accident Conditions*, Columbus, Ohio, Oct. 28-Nov. 1, 1985, International Atomic Energy Agency, Vienna, Austria, March 1986.

13. G. W. Parker, "The Chemistry of Nuclear Reactor Accidents: Problems and Progress," presented at ACS National Meeting, Sept. 8-12, 1986, Anaheim, Calif.; to be published in *Proceedings on Chemical Phenomena Associated with Radioactivity Releases During Severe Nuclear Plant Accidents*.

(1) initial release into the containment, (2) interaction of iodine species in the containment not directly involving water pools, (3) interaction of iodine species in or with water pools, and (4) special interactions.

The formation of volatile iodine as I_2 or organic iodides is dependent on radiation and solution pH. Lower pH levels result in increased formation of volatile iodine species; thus a pH of 3.05 results in a conversion of I^- to I_2 , which is more than two orders of magnitude greater than that for tests run at a pH of 6.1 or 6.8.

Iodine in water pools may interact with impurities in the water to produce chemical compounds or oxidation states that can alter the volatility of the iodine. Silver from pressurized-water reactor (PWR) control rod alloy could play an important role in determining the ultimate chemical form of iodine. Silver in the presence of radiation has proven to be effective in converting dissolved iodine species into solid, insoluble silver iodide and thereby decreasing iodine volatility.

Inorganic iodide species can react with a wide variety of organic compounds to produce organic iodides. In addition to straightforward reaction of organic compounds and iodine species, radiation in the containment could bring about free radical reactions not otherwise possible.

The formation of organic iodides within or above water pools has been shown to be dependent on (1) the presence of iodine as I_2 , (2) radiation, and (3) organic material. Tests performed in the cobalt gamma irradiator showed that CH_3I formed predominantly in the aqueous phase and that there was a correlation between iodine as I_2 and greater conversion to CH_3I .

Another interesting result of these studies is that the CH_3I must have formed predominantly in the aqueous phase rather than in the gas phase. The organic iodide partition coefficients observed in these tests showed that more CH_3I was present in the aqueous phase than would be expected in an equilibrium gas-aqueous partitioning. Thus, for example, tests run at pH 6.0 in 0.2 mol/dm³ borate solution had a CH_3I partition coefficient (defined as concentration of CH_3I in aqueous solution divided by concentration of CH_3I in gas) of 270, and the equilibrium partition coefficient at the 30 to 35°C temperature used in these tests would be ~2 to 3. Therefore, the CH_3I must have formed predominantly in the aqueous phase and was then stripped to the gas phase.

A model called TRENDS (Transport and Retention of Nuclides in Dominant Sequences) has been structured to follow the distribution of iodine in the containment and to calculate the amount released during severe accidents. Chemical forms of iodine used in this code include I_2 , HI, CH_3I , CsI, I^- , and AgI. These forms can be distributed in the gas, in water, on aerosols, on painted surfaces, on metal surfaces, and on concrete surfaces and can be vented or leaked out. Both physical transfer between phases or locations and chemical transfer by reaction are considered in the model. Thus, for example, I_2 can be distributed physically in the gas, in water, and as deposited on aerosols, steel, concrete, or paint. In addition, I_2 can react to form CH_3I , I^- , or AgI.

A version of the TRENDS code has been used in a special limited study in support of NUREG-0956¹⁴ to evaluate the influence of chemical processes within the containment on the iodine source term when the chemical form released from the reactor coolant system was CsI and/or HI. The purposes of this study, then, were (1) to evaluate the impact on the iodine source term of the potential conversion of CsI into "volatile" iodine within the reactor coolant system and (2) to evaluate the general influence on the iodine source term of heretofore neglected chemistry and transport phenomena in containment.

Tellurium Behavior in Containment. The initial chemical forms of tellurium transported to the containment are likely to be metal tellurides such as SnTe or Cs₂Te, hydrogen telluride (H₂Te), elemental tellurium, and perhaps some organic tellurides. These initial chemical forms may be altered by oxidation or by formation of organic tellurides. The chemical systems of interest for the reactions of tellurium species include (1) the fission product material released from the primary system, (2) steam, (3) liquid water, (4) air, (5) organic material, and (6) metal and concrete surfaces. In addition, these materials will be in radiation fields of variable intensities.

Cesium telluride and hydrogen telluride are the only chemical forms of tellurium likely to be readily soluble in water, and both of these compounds decompose in water to precipitate elemental tellurium. The precipitated tellurium can then

14. M. Silberberg, J. A. Mitchell, R. O. Meyer, and C. P. Ryder, *Reassessment of the Technical Bases for Estimating Source Terms*, NUREG-0956, U.S. Nuclear Regulatory Commission, July 1986.

easily oxidize to TeO_2 in a water-air system. Thus the principal dissolved tellurium species still remaining will result from the limited solubility of tellurium dioxide.

Of the three volatile tellurium chemical forms [Te_2 (gas), H_2Te , and organic tellurides], only the latter have the potential to remain in the gas phase in a containment atmosphere. The Te_2 (gas) would rapidly condense and oxidize, and the H_2Te would decompose to elemental tellurium. There may be some transport of these inorganic forms on initial release into the containment; however, they should be dissipated in a short, but indeterminate, time. The extent and composition of organic telluride formation under containment conditions are largely unknown. Tellurium reacts with organic free radicals to produce organic tellurides, which, in turn, may oxidize to yield organic telluroxides. Organic tellurides are not as volatile as organic iodides; but, since they are insoluble in water, they will not partition between the aqueous and the gas phase.

Iodine Release from a Steam Generator Tube Rupture. A tube failure within the steam generator of a PWR results in the escape of primary coolant into the secondary system. The release of primary coolant from a steam generator tube rupture is a design-basis accident; and, for licensing purposes, conservative assumptions are made to estimate the release of radioactivity. A better understanding of iodine behavior would help to establish the margin of conservatism.

Iodine partition coefficients (concentration of iodine species in the aqueous solution divided by the iodine concentration in the vapor phase) have been measured at simulated PWR steam generator conditions (285°C and 6.9 MPa) using carrier-free radioactive ^{131}I as sodium iodide. The iodine tracer concentration has been maintained at about 6×10^{-11} mol/L. The boric acid concentration has been varied from 0 to 0.4 mol/L, and the solution pH (measured at 25°C) has been adjusted from 4 to 9 by the addition of lithium hydroxide. The simulated steam generator is an 8.9-cm-diam, 152-cm-long, bolted-closure autoclave fabricated of type 316 stainless steel and heated by three external furnaces with separate power controls.

As might be expected from iodine species measurements in a PWR primary coolant, iodine partition coefficients decreased with increasing boric acid concentration; however, the iodine volatility is essentially independent of the solution pH for a given boric acid concentration. Sparging the solu-

tions with air at room temperature (before heatup to test temperature) increases the iodine volatility by an order of magnitude compared with argon sparging. Iodine partition coefficient measurements, in grams per cubic centimeter of liquid per grams per cubic centimeter of vapor, ranged from a low of 200 (0.2 M boric acid sparged with air) to 400,000 (purified water sparged with argon). Iodine volatility behavior was comparable in either 0.002 M boric acid solutions or distilled and deionized water.

The iodine partition coefficient value used in the design-basis accident calculation for a PWR steam generator tube rupture is $\sim 2,000$ in concentration units at 285°C . In 60% of the runs completed (31 of 52), the solutions were prepared and handled in air with no additional gas sparging. With these conditions, the only iodine partition coefficient measurement below 2,000 was in a 0.4 M boric acid solution.

TRAP-MELT Validation Program

Calculational results published as part of the Battelle-Columbus Source Term Study¹⁵ indicate that, for some LWR severe accident sequences, aerosol deposition in the reactor coolant system can lead to significant reductions in the radionuclide release to the environment. For the Battelle study, aerosol transport and deposition in the reactor coolant system were calculated using the TRAP-MELT2 computer code, which was developed at Battelle. The objective of the ORNL TRAP-MELT Validation Program, which is sponsored by the Accident Evaluation Division of the NRC, is to conduct simulated reactor-vessel upper-plenum aerosol deposition and transport tests. The results from these tests will be used in the ongoing effort to validate TRAP-MELT2. The project includes two experimental subtasks: the Aerosol Transport Tests and the Aerosol Resuspension Tests.

In the Aerosol Transport Tests, aerosol transport in a vertical pipe has been studied; this geometry was chosen to simulate aerosol deposition and transport in the reactor-vessel upper plenum. Tests A105 to A108, the final four of the planned tests, were completed. In these tests the configuration illustrated in Fig. 7.3 was used. Aerosols were generated by feeding metal powder to a plasma torch

15. J. A. Gieseke et al., *Radionuclide Release Under LWR Accident Conditions—Volumes I–VII (Draft)*, BMI-2104, Battelle-Columbus Laboratories, 1983–1984.

Fig. 7.3. Aerosol transport test section components and measurement locations.

aerosol generator. In the tests, either iron oxide aerosols were made by oxidizing iron metal powder, or metallic zinc aerosols were made by direct vaporization of zinc metal powder. The generated aerosols can agglomerate, deposit on the pipe sidewalls, or be transported out of the pipe into an aerosol collection bag. Results from these tests were summarized in a series of letter reports¹⁶⁻¹⁹ and in a recently published book.²⁰

For tests A105 and A106 (iron-oxide experiments), the test data were compared with TRAP-MELT2 calculations.²¹ As in the analyses performed for tests A103 and A104 (also performed with iron oxide), the results of the A105 and A106 comparisons were not satisfactory in that the code

16. A. L. Wright and W. L. Pattison, *Aerosol Transport Test A105 Preliminary Data Report*, ORNL/NRC/LTR-85/22, September 1985.

17. A. L. Wright and W. L. Pattison, *Aerosol Transport Test A106 Preliminary Data Report*, ORNL/NRC/LTR-85/30, December 1985.

18. A. L. Wright and W. L. Pattison, *Aerosol Transport Test A107 Preliminary Data Report*, ORNL/NRC/LTR-86/2, June 1986.

19. A. L. Wright and W. L. Pattison, *Aerosol Transport Test A108 Preliminary Data Report*, ORNL/NRC/LTR-86/10, August 1986.

20. A. L. Wright and W. L. Pattison, "Results from Simulated Upper-Plenum Aerosol Transport Tests," pp. 1110-13 in *Aerosols and Reactivity*, Pergamon Press, 1986.

21. A. L. Wright and W. L. Pattison, *Summary of TRAP-MELT2 Results for Aerosol Transport Tests A105 and A106*, ORNL/NRC/LTR-86/9, December 1986.

significantly—by at least a factor of 8—under-predicted the measured aerosol wall plateau.

The objective of the Aerosol Resuspension Tests is to provide a data base for developing resuspension-rate models that can be included in TRAP-MELT2 and other aerosol transport codes. This work is being done because none of the present aerosol transport codes include resuspension models. The "Series-2" resuspension tests were recently completed; the test apparatus for these experiments is illustrated in Fig. 7.4. Aerosols of manganese oxide, tin oxide, iron oxide, manganese, or zinc were first generated and deposited on the interior of a series of 2.5-cm-diam, 7.6-cm-long steel tubes [Fig. 7.4(a)]. The tubes were then mounted in a second test section [Fig. 7.4(b)] in which the actual resuspension measurements were made. Nitrogen was used as the flow gas in these tests, and resuspension measurements were made at flow velocities ranging from 4 to 60 m/s. The results from the experiments are now being evaluated.

LACE Code-Experiment Coordination

A major area of uncertainty in the evaluation of radioactive source terms from LWR severe

accidents is the level of validation of the aerosol transport computer codes used to calculate the source terms. A number of aerosol transport computer codes are used throughout the world to predict the behavior of aerosols in the reactor coolant system and the reactor containment. The models used in these codes must be validated against experimental results. The LWR Aerosol Containment Experiment (LACE) Code-Experiment Coordination Project (funded by the Electric Power Research Institute) is the most comprehensive international aerosol code validation effort that has ever been undertaken. Six large-scale aerosol transport experiments have been performed at the Hanford Engineering Development Laboratory as part of the internationally funded LACE program. The efforts at ORNL consist of (1) providing guidance to participating code analysts for performing calculations to model the tests, (2) compiling the test and calculational results, and (3) critically evaluating comparisons of code results to the test data. An international group of aerosol code analysts from Finland, Germany, Japan, Italy, Sweden, the United Kingdom, and the United States is performing calculations to predict the experimental results.

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(a) APPARATUS FOR COLLECTING DEPOSITION SAMPLES (WILL BE ORIENTED VERTICALLY):

(b) RESUSPENSION TEST APPARATUS:

Fig. 7.4. "Series-2" aerosol resuspension test equipment.

Code comparison results from the first three tests have been documented.^{22,23} Results of code-data comparisons for the first LACE experiment, LA1, were transmitted to project participants. For this test, 29 aerosol transport calculations were performed, using nine different aerosol codes, by analysts from six countries. This effort represents a significant improvement over all past aerosol code comparison efforts for two reasons: (1) each code calculation and the code inputs used for each calculation were investigated in detail so that we could determine why the different codes produced different results and (2) the errors in the calculated values for each important aerosol transport parameter were calculated and clearly presented. The main findings of this effort can be summarized as follows: (1) most codes calculated the important aerosol transport parameters within a factor of 2 and (2) uncertainties in thermal-hydraulic code input parameters, particularly those related to when steam condensation would occur, had a major influence on the calculated aerosol transport results. Perhaps, however, the major outcome of the efforts to date has been that all the aerosol code users have improved in their ability to use the various codes.

7.2 HIGH-LEVEL-WASTE REPOSITORY LICENSING SUPPORT

This project provides the NRC with technical assistance in the analysis and evaluation of geochemical information relevant to the candidate high-level-waste geologic repository sites being characterized by DOE. Technical assistance involves participation in site visits and data reviews, preparation of written technical commentary and reviews, and organization of peer reviews. Major activities during this report period included the following:

1. review of the draft environmental assessment reports issued by DOE for three candidate repository sites—Yucca Mountain, Nevada; the Hanford Site, Washington; and Deaf Smith County, Texas;
2. analysis of the radionuclide solubility data base and the information descriptive of geochemical conditions at the three candidate sites; and
3. a peer review workshop that addressed the methodology for obtaining radionuclide sorption information.

The workshop identified concerns relative to the methodology for characterizing and modeling sorption processes in geologic sites that might be used as high-level-waste repositories. Generally, current practice is to use a simplified sorption model (sometimes called the K_D method²⁴) to describe radionuclide migration in rock-groundwater systems. The K_D method, which is based on chromatographic theory, was developed for saturated, porous, ion-exchange media. In this method, the retardation of solutes relative to the flow of solution is described by the expression

$$R_F = 1 + K_D(\sigma/\phi), \quad (1)$$

where

R_F = the retardation factor,

σ = bulk density,

ϕ = porosity,

K_D = an experimentally measured equilibrium distribution coefficient.

Predictions of the mobilities of some radionuclides in geologic systems may prove to be inaccurate because sorption in the rock-groundwater systems may be poorly represented by this linear model. Specifically, in geologic systems, the underlying theoretical concept, the data measurement techniques, and the mathematical modeling used in the K_D method are subject to considerable uncertainty.

In a high-level-waste repository, several cubic kilometers of rocks will surround the emplaced waste. In addition to physical isolation from the environment, groundwater release pathways through this large mass of rock may provide, for some sites, an extensive sorptive medium for the retardation of solubilized radionuclides. The NRC and the U.S. Environmental Protection Agency (EPA) have established regulations for the release

22. A. L. Wright, J. H. Wilson, and P. C. Arwood, "Pretest Aerosol Code Comparisons for LWR Aerosol Containment Tests LA1 and LA2," pp. 1114-17 in *Aerosols and Reactivity*, Pergamon Press, 1986.

23. A. L. Wright, J. H. Wilson, and P. C. Arwood, "Summary of Aerosol Code-Comparison Results for LWR Aerosol Containment Tests LA1, LA2, and LA3," to be published in the proceedings of the Fourteenth Water Reactor Safety Meeting, Gaithersburg, Md., October 1986.

²⁴The distribution coefficient, K_D , is defined as the ratio of the concentration of the sorbed element on the solid divided by the concentration in the solution. The units are liters per kilogram.

of radionuclides from high-level-waste repositories. The EPA standard²⁴ specifies cumulative release limits for radionuclides over a 10,000-year period at a distance of 5,000 m from the repository. The NRC regulation²⁵ specifies performance criteria for the repository engineered systems and also states that the EPA standard must be met. The environmental assessments for the Yucca Mountain²⁶ and the Hanford Site²⁷ candidate repositories include an analysis of the retardation of solubilized radionuclides in groundwater by sorption onto the tuff at Yucca Mountain and the basalt at the Hanford Site. The mobilities of many radionuclides relative to groundwater movement were calculated to be retarded by orders of magnitude as a result of sorption reactions. These predictions were based on application of the K_D method.

Important considerations in the application of the K_D method to geologic systems such as repository sites are discussed in the following paragraphs.

- In most experimental determinations of K_D , sorption data are obtained using gram-scale amounts of pulverized whole-rock samples. Water/rock ratios typically range between 5 and 10. Application of the K_D data to repositories requires some method of extrapolation to the geologic conditions in the host rocks of the repositories. Use of Eq. (1) implies that a simple linear method of extrapolation is applicable because the units of K_D are liters per kilogram (i.e., the extent of sorption is proportional to the total mass of the rock). This method of extrapolation is valid only if (1) the entire mass of rock in the repository participates in the sorption process and (2) groundwater flow is uniform

throughout the far field of the repository system. In geologic systems, the occurrence of fracture flow of groundwater, slow diffusion of radionuclides into rocks, slow sorption reaction kinetics, and groundwater flow in unsaturated regimes severely complicate application of the K_D approach.

- Even at low concentrations of a radionuclide, the measured K_D value may not be independent of the radionuclide concentration; therefore, sorption cannot be accurately modeled by a single K_D value, as in Eq. (1). At higher radionuclide concentrations, the sorption sites could become saturated; at this point, sorption would be nil, and Eq. (1) would be completely inapplicable.
- Radionuclides may undergo chemical reactions during migration, may be present as multiple species, and may compete with other radionuclides or groundwater constituents for available sorption sites. Equation (1) does not consider such reactions or competition.
- The applicability of Eq. (1) is further reduced if radionuclides migrate as colloids or are adsorbed onto groundwater colloidal material.

24. U.S. Environmental Protection Agency, 40 CFR Part 191, *Environmental Standards for the Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Wastes*, Washington, D.C., 1985.

25. U.S. Nuclear Regulatory Commission, 10 CFR Part 60, *Disposal of High-Level Radioactive Wastes in Geologic Repositories*, Washington, D.C., 1986.

26. U.S. Department of Energy, *Environmental Assessment, Yucca Mountain Site, Nevada Research and Development Area, Nevada*, DOE/RW-0073, Washington, D.C., May 1986.

27. U.S. Department of Energy, *Environmental Assessment, Reference Repository Location, Hanford Site, Washington*, DOE/RW-0070, Washington, D.C., May 1986.

8. CONSOLIDATED EDISON URANIUM SOLIDIFICATION PROJECT

During the early 1960s, the U.S. Atomic Energy Commission (AEC) sponsored a test irradiation of a $\text{ThO}_2\text{-UO}_2$ fuel core in the Consolidated Edison Indian Point-1 Reactor at Indian Point, New York. The test generated ~1000 kg of highly radioactive and fissile uranium (containing ~75% ^{235}U , ~10% ^{233}U , and ~140 ppm ^{232}U). In 1968, the AEC also sponsored reprocessing of the irradiated fuel at the Nuclear Fuel Services Plant in West Valley, New York, to recover the fissile uranium. The uranium nitrate product solution resulting from the reprocessing operations was sent to ORNL for storage. Soluble poisons (cadmium nitrate and gadolinium nitrate) were added to ensure subcriticality of this solution during its storage in a specially prepared 18,900-L (5,000-gal) Thorium Reactor Uranium Storage Tank (TRUST), located at the Radiochemical Processing Plant (RPP).

When no use for the uranium had become apparent after several years of storage, a project called the Consolidated Edison Uranium Solidification Program (CEUSP) was initiated in the late 1970s to solidify the material for safe long-term storage. A unique conversion/solidification process and the equipment to carry it out were conceived, developed, and operated by Chem Tech personnel.¹⁻³ The process equipment was designed by personnel in the Engineering Division of Martin Marietta Energy Systems, Inc., fabricated by members of the Plant and Equipment Division of

ORNL, and installed in Cells 3 and 4 of the RPP by employees of Rust Engineering Company.⁴ Quality inspections were done by the ORNL Quality Department, and process instrumentation was installed by ORNL Instrumentation and Controls Division personnel. During testing and operation of the CEUSP Facility, analytical chemistry services were provided by members of the ORNL Analytical Chemistry Division.

Installation and testing of the process equipment and instrumentation and approval of the safety analysis and quality assurance documentation were completed in early 1985. The final readiness reviews were completed in April 1985, and operation of the CEUSP Facility began at that time.

8.1 SOLIDIFICATION OPERATIONS

A schematic diagram of the process flowsheet is shown in Fig. 8.1. The material was processed in batches—each containing ~2.6 kg of uranium. The key process steps included (1) a batch evaporation to concentrate the uranium-cadmium-gadolinium nitrate solution to slightly less than the saturation concentration; (2) the use of formaldehyde in the evaporator to destroy nitric acid so that crystallization of the nitrate salts due to supersaturation would not occur; (3) the use of an in-situ thermal denitration process to convert uranium, cadmium, and gadolinium to the oxide form inside the storage cans; (4) the use of remotely operated equipment to weld lids onto the storage cans; (5) encapsulation of each can within a crimp-sealed, secondary containment canister; and (6) transfer of the finished packages to a shielded storage well.

1. C. P. McGinnis, E. D. Collins, R. Hall, J. K. Johnson et al., "Development and Operation of a Unique Conversion/Solidification Process for Highly Radioactive and Fissile Uranium," in press.

2. R. J. Vedder, E. D. Collins, and P. A. Haas, "Development of the CEUSP Thermal Denitration In-Storage Can Process," *Proc. Symp. Waste Management '86*, R. G. Post, ed., Univ. of Arizona, Tucson, 1986, Vol. 1, pp. 309-12.

3. R. Hall, B. D. Patton, and P. A. Haas, "Reaction of Formaldehyde and Nitric Acid in a Remotely Operated Thermosiphon Evaporator," *Proc. Symp. Waste Management '86*, R. G. Post, ed., Univ. of Arizona, Tucson, 1986, Vol. 1, pp. 313-16.

4. C. P. McGinnis, E. D. Collins, and B. D. Patton, "A Remotely Operated Facility for In-Situ Solidification of Fissile Uranium," *Proc. Symp. Waste Management '86*, R. G. Post, ed., Univ. of Arizona, Tucson, 1986, Vol. 1, pp. 453-56.

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Fig. 8.1. Process flowsheet for the Consolidated Edison Uranium Solidification Program (CEUSP) Facility.

The evaporation step, in which the feed solution was concentrated by a factor of ~ 2.5 , was included in the process design to reduce the overall time requirement for the solidification process. Including the evaporation step made it necessary to add formaldehyde as an agent for acid destruction; otherwise the concentrated solution produced by the evaporation would have been supersaturated, potentially causing solids formation and equipment fouling. Since the evaporation step of the process required only 2 to 3 h while the thermal denitration required ~ 24 h, the CEUSP Facility included three parallel thermal denitration systems and only one evaporation system.

The process equipment was located in enclosures that were carefully contained to prevent release of the radioactive materials and were heavily shielded

to protect the operating personnel from high-energy gamma rays. Most of the equipment was controlled remotely by means of instruments operated from an adjoining control room, although some equipment, such as the storage can conveyor and grapple, were operated manually using crankwheels located outside of the shielding walls.

Direct, hands-on maintenance of the process equipment was required, because the constraints of the available facility did not permit remote maintenance. Maintenance-susceptible pieces of equipment (e.g., valves, pumps, and furnaces) were located in cubicles and were made accessible via removable shielding and doors.

During preoperational checking of the mechanical equipment, 10-year life-cycle tests were performed on many of the components to select durable materials that would most likely require infrequent maintenance. Several components failed this rigorous testing, and more reliable substitutes were found. For the components considered likely to require repair or replacement (such as the denitration feed pumps), maintenance procedures were devised and tested under simulated operating conditions. These procedures included the erection of containment tents, the use of special ventilation methods, and the dressout of maintenance personnel in masks and double layers of protective clothing.

After actual operations began, only four shutdowns were required. During each shutdown, all three denitration pumps were replaced. Although minor problems occurred, the repairs were successful and the maintenance personnel received only insignificant radiation exposures. This success was attributed both to the procedures that had been developed and tested prior to initial operation and to the skill of the craftsmen.

Approximately 400 batches of uranium were solidified and stored during the CEUSP Facility operation, which was successfully completed in June 1986, nearly 2 months prior to the scheduled completion date (Fig. 8.2).

8.2 CLEANOUT OF THE CEUSP FACILITY

Following solidification of the uranium that had been produced in the Consolidated Edison Indian Point-I Reactor, the CEUSP Facility was used to

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Fig. 8.2. Comparison of the actual CEUSP processing rate with the scheduled rate.

solidify ~65 kg of ^{233}U containing 20 ppm of ^{232}U . Final cleanup of the CEUSP Facility and the TRUST was then begun. The internal surfaces of the TRUST were thoroughly cleaned by filling the tank with dilute nitric acid, followed by mixing

and soaking for an extended time. Recoverable amounts of uranium were solidified and stored. The cleanup operations should be completed and the CEUSP Facility should be placed in standby condition by July 1987.

9. RADIOACTIVE MATERIALS PRODUCTION

The Office of Nuclear Materials Production (ONMP) of the U.S. Department of Energy sponsors two major programs in the Chemical Technology Division for production of radioactive materials. These are the ^{233}U Storage, Processing, and Distribution Program and the ^{252}Cf Industrial Sales/Loan Program.

9.1 URANIUM-233 PROCESSING

The Radiochemical Processing Plant (RPP) at ORNL has served for more than two decades as the national repository and distribution center for ^{233}U . Several hundred kilograms of uranium, containing approximately 90 to 98% ^{233}U , are stored at the RPP in the form of metal, oxides, and nitrate solutions. All of these uranium materials contain small, but significant, concentrations of ^{232}U , ranging from 2 to 225 ppm. Most of the radioactivity associated with the ^{233}U comes from decay daughters of the impurity, ^{232}U (74-year half-life). Fully contained storage and handling facilities are required because the daughters are predominantly alpha emitters. In addition, three decay daughters, ^{228}Th (1.9-year half-life), ^{220}Rn (54.5-s half-life), and ^{208}Tl (3.1-min half-life), require special handling.

The ^{208}Tl emits a 2.6-MeV gamma ray, which necessitates that the ^{233}U storage and processing facilities must be heavily shielded and remotely operated. The ^{220}Rn daughter is a gas, which requires the RPP to have special off-gas treatment facilities to retain the off-gas streams long enough to permit ^{220}Rn decay; high-efficiency filtration equipment is also necessary to remove the particulate decay daughters of the ^{220}Rn . Since these decay products include ^{208}Tl , the filters must be either shielded or remotely located.

The ^{228}Th daughter of ^{232}U decays to produce short-lived radionuclides (including ^{220}Rn and ^{208}Tl); thus, by chemically separating thorium from the uranium materials, the gamma-emitting daughters can be removed temporarily from the

^{233}U . Decontamination can be achieved to reduce the radiation by factors greater than 100 for several days and by factors greater than 50 for approximately 1 month. As time passes the concentrations and the associated radiation emissions of the ^{232}U daughters again increase.

The RPP contains chemical processing equipment that is used (1) to dissolve multikilogram batches of U_3O_8 , UO_2 , or $\text{ThO}_2\cdot\text{UO}_2$; (2) to separate ^{228}Th and its daughters from the resulting uranyl nitrate solutions by solvent extraction and ion exchange; and (3) to convert the purified uranium to oxides (U_3O_8 or UO_2). The equipment used for the dissolution and separation processes is located in heavily shielded hot cells and is remotely operated (but directly maintained). The conversion equipment is located in an unshielded glove-box facility; therefore, only purified ^{233}U containing less than 10 ppm of ^{232}U can be processed in the facility without exposing the operating personnel to unacceptable gamma radiation.

Storage facilities for ^{233}U at the RPP include ten 400-L tanks for liquid storage, shielded storage wells for solids, and a safeguarded vault for small packages and samples. Until early 1985, the vault was used for storage of multikilogram amounts of ^{233}U . To improve safeguards, the inventory of special nuclear materials in the vault was reduced significantly by transferring most of the materials to storage in the shielded wells. Massive cover blocks and locking plates were installed over the shielded wells so that theft cannot be accomplished without a very significant time delay.

In recent years, receipts and shipments of ^{233}U have decreased substantially. Therefore, in 1984 the ONMP instructed ORNL to solidify all ^{233}U and to clean out the processing facilities and place them in a long-term standby condition. This work is scheduled for completion in FY 1989.

Between April 1985 and December 1986, four small batches of ^{233}U , ranging in size from 250 g to 3.1 kg, were purified, converted to U_3O_8 , and shipped to Atomic Energy of Canada, Limited.

Also, 25 kg of ^{233}U was purified, converted to U_3O_8 , and stored for subsequent shipment to Los Alamos National Laboratory. Preparation of a similar amount for subsequent shipment to Lawrence Livermore National Laboratory is in progress. In addition, ~ 65 kg of ^{233}U (containing ~ 20 ppm of ^{232}U) was solidified by thermal denitration in the CEUSP Facility (see Sect. 8) and placed in the storage wells.

9.2 CALIFORNIUM-252 INDUSTRIAL SALES/LOAN PROGRAM

The ^{252}Cf Industrial Sales/Loan Program, which is sponsored by the ONMP, involves loans of ^{252}Cf neutron sources to agencies of the U.S. government and sales of ^{252}Cf as the bulk oxide and as palladium-californium alloy pellets and wires. The program was operated from 1968 through 1986 in temporary facilities at the Savannah River Laboratory (SRL). These program activities were

transferred in 1986 to the TURF Californium Facility (TCF) in Bldg. 7930, and the hot-cell facilities at SRL are now being decommissioned.

Since 1970 the staff of the ORNL Transuranium Processing Plant (TPP) has operated the TCF as part of the Heavy-Element Research Program sponsored by the DOE Office of Basic Sciences (BES). The facility was built to provide hot-cell space that is free of ^{244}Cm contamination in which to recover very pure ^{248}Cm , the alpha decay daughter of ^{252}Cf , for use in research. Since operation began, the TCF has processed essentially all of the ^{252}Cf used in the Western world, including that used in the Industrial Sales/Loan Program.

In 1983 the ONMP made a decision to transfer the ^{252}Cf Industrial Sales/Loan Program to the TCF, and DOE authorized the addition of facilities and equipment to accommodate the program's activities while continuing the BES Heavy-Element Research Program work. The expanded TCF (see Fig. 9.1) has the capability for producing and

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Fig. 9.1. Schematic diagram of the TURF Californium Facility following modifications made to accommodate the ^{252}Cf Industrial Sales/Loan Program activities.

packaging bulk californium, as well as fabricating neutron sources in all of the physical forms required for both the research and industrial sales/loan programs.

The original TCF was located entirely within Cell G, which is highly contaminated with ^{252}Cf . All processing and fabrication activities involving unencapsulated ^{252}Cf are still performed there. The cell contains two identical sets of ion-exchange equipment for separating curium from ^{252}Cf by means of elution development chromatography, using α -hydroxyisobutyrate as the eluent. One set of equipment is used to remove residual curium from the ^{252}Cf as soon as it is received, while the other set is used to recover the daughter product, ^{248}Cm , from ^{252}Cf that has been stored ~ 2 years.

For the Heavy-Element Research Program, a solid form of ^{252}Cf is produced by sorbing it onto cation-exchange resin and calcining the resin to form small spheres of californium oxysulfate. The spheres are the final form used for storage and bulk shipment; they are blended with aluminum powder and pressed into pellets for fabricating research neutron sources.

For the Industrial Sales/Loan Program, the ^{252}Cf is converted to a variety of solid forms for bulk shipment and neutron source fabrication. The primary processes common to the production of all of the solid forms are oxalate precipitation followed by calcination to the oxide. The pure Cf_2O_3 is used for many bulk shipments and neutron sources. Also, a palladium-californium alloy (or cermet) may be formed by adding palladium dur-

ing the oxalate precipitation step, pressing the palladium-coated Cf_2O_3 into pellets following calcination, and heating the pellets. These pellets, or palladium-californium wires formed by processing the pellets through a rolling mill, are the final forms for other shipments and neutron sources.

The processing and fabrication steps are performed using the rolling mill and a variety of small processing vessels, furnaces, hydraulic presses, and dies that are located conveniently throughout Cell G. This hot cell also contains equipment for welding, helium leak-testing, X-ray examination, decontamination, and assaying the ^{252}Cf content of the bulk shipments and neutron source capsules.

New work stations were installed in adjoining Cell C (Fig. 9.1) for welding secondary capsules, examining welds by X-ray and by helium leak-testing, smear-testing for surface contamination, californium assay by neutron counting, final decontamination, and loading of large shipping casks. Cell B contains a work table for loading and unloading small shipping casks. Since all ^{252}Cf handled outside Cell G is encapsulated, Cells B and C remain uncontaminated; personnel enter these areas routinely for shipping cask setup, equipment maintenance, and waste removal. A californium storage facility was installed in the water-filled basin originally designed for the unloading of fuel casks.

The work areas are interconnected by pneumatic systems that are used to transfer neutron sources and packages from one hot cell area to another and to the californium storage facility.

10. FISSION ENERGY

10.1 CONVERSION PROCESSES FOR BREEDER REACTOR FUEL MATERIALS

Members of the ORNL Chemical Technology Division and fabrication development personnel at the Pacific Northwest Laboratory (PNL) and at the Hanford Engineering Development Laboratory (HEDL) have participated in a joint project managed by the national Consolidated Fuel Reprocessing Program (CFRP) to develop advanced processes for recycling uranium and plutonium fuels for breeder reactors.

An assessment of various conversion processes was done for this project, including both a technical evaluation and a cost/benefit analysis. Results indicated that the DIPRES (Direct PRESSpheroidized) process, developed at ORNL, appeared to be the most favorable for both process and product improvement.

The Chem Tech area of responsibility for this development program was the preparation of plutonium-uranium oxide microspheres, which were subsequently pressed into fuel pellets that were used in the fabrication of fuel pins for an irradiation test in the Fast Flux Test Facility (FFTF) at HEDL. The 25 wt % PuO_2 -75 wt % UO_2 spheres were prepared at ORNL by an internal gelation process. Since the work was completed during this report period, the gel-sphere preparation facility is now being maintained in standby condition. Using the oxide spheres, fuel pellets were formed by the DIPRES process, and 27 test pins were fabricated and inspected at HEDL. The pins were loaded into the FFTF for irradiation late in 1986 and are expected to be removed for postirradiation examinations in late 1988.

A backup conversion process for uranium and plutonium fuels, also developed at ORNL, is modified direct denitration (MDD). This process has the advantages of simple waste treatment and the same small number of process steps as the thermal denitration of uranium nitrates. The modified

chemical flowsheet produces a UO_3 or $\text{UO}_3\text{-PuO}_2$ powder having excellent ceramic characteristics for the fabrication of reactor fuel pellets.¹

Procurement and design activities are under way for a project using the MDD process in a rotary kiln at ORNL for in-cell preparation of ^{233}U oxides from nitrate solutions. Division researchers have also supplied information and assistance for an evaluation of MDD as a conversion process in an aqueous flowsheet for the purification of uranium enriched by the Atomic Vapor Laser Isotope Separation (AVLIS) method. This led to a proposal for a large rotary kiln to demonstrate MDD in an AVLIS product-conversion pilot plant.

10.2 FFTF FUEL DISSOLUTION STUDIES

A series of studies to characterize spent fuels as a function of burnup was also completed during this period. Details of the results cannot be given in this report because DOE regulations stipulate that such information must be distributed only as applied technology (Category UC-86T). The general directions of the work are indicated here.

Fuel from the FFTF at Richland, Washington, with a peak burnup of ~90,000 MWd per metric ton of initial heavy metal (MTIHM), was sheared and used in dissolution tests. Intact pins were gamma-scanned to determine the longitudinal distribution of various isotopes; significant migrations of cesium and antimony were observed. The shear force required for a single-pin shear was determined as a function of position along the pin. Required shear forces near the center of the pins were found to be lower than those for shearing the ends of the pins, where burnup is lower. Much more fuel was dislodged from the pins when high-burnup fuel was sheared, and the dislodged pieces

¹P. A. Haas and W. B. Stinea, "Method for Improved Decomposition of Metal Nitrate Solutions," U.S. Patent 4,409,157, October 11, 1983.

were much larger than those observed with fuel of lower burnup. The cladding was also found to be more brittle, and it fragmented to a greater extent during shearing than that for fuel of lower burnup. Our dissolution studies examined several features: (1) the quantity of residue remaining after dissolution of fuel samples in nitric acid, (2) the quantity of plutonium recovered from the residue by leaching with HNO_3 -HF solutions, (3) the fractions of the expected uranium and plutonium recovered during dissolution, and (4) the behavior of various fission products during dissolution.

10.3 SETF STUDIES

The Solvent Extraction Test Facility (SETF) is located in one of the heavily shielded hot cells of the Transuranium Processing Plant (TPP). The SETF, which first became operational in late 1978, was used to study modified Purex flowsheets for the reprocessing of both light-water reactor (LWR) and, most recently, fast-breeder reactor (FBR) fuels. This work was supported through FY 1986 by the CFRP. The tests provided information on heavy metal recoveries, fission product behavior, and general operability of the system, which established a basis for comparing flowsheet options. Also, in-line instrumentation for process control was evaluated.

The processing steps that were done in the SETF included dissolution of the fuel in nitric acid, clarification and adjustment of the dissolver solutions, solvent extraction, nitrate-based anion exchange, and plutonium oxalate precipitation followed by calcination. The solvent extraction contactors consisted of three banks of 16-stage mixer-settlers capable of processing several kilograms of heavy metals per day. However, the solvent extraction tests were limited to less than 1 d of continuous operation because of safeguard restrictions limiting the amount of plutonium that could be handled in each test.

During this report period, four solvent extraction tests were completed using FFTF-irradiated fuel samples having burnups of $\sim 90,000$, $\sim 60,000$ (average), and $\sim 55,000$ MWd/MTIHM and cooling times of 1, 2-5, and 3 years, respectively. The organic extractant for these tests was tri-*n*-butyl phosphate (TBP) diluted with a normal paraffin hydrocarbon to make either a 10 vol % or a 30 vol % solution.

Two different partitioning techniques were investigated for separating uranium and plutonium. One technique, similar to the traditional Purex approach, used 30 vol % TBP solvent and reduced the plutonium to the relatively inextractable Pu(III) valence with a chemical reductant, hydroxylamine nitrate. The other method used no reductant and relied entirely on the different extractabilities of the U(VI) and Pu(IV) complexes. In order to enhance this difference and improve the separation, these tests were made using a relatively low (10 vol %) TBP concentration and lower operating temperatures than are traditionally used in Purex flowsheets. During the final two tests, stable operation of the coextraction-coscrub contactor was successfully maintained by means of a computerized control system. An in-line sampling loop, equipped with a spectrophotometer, was used to measure the uranium and plutonium concentrations from a selected stage in the extraction section. Based on these data, the control system automatically adjusted the extractant addition rate to achieve high solvent loadings while maintaining low losses to the aqueous raffinate.

10.4 PLUTONIUM CHEMISTRY AND NEW FLOWSHEET CONDITIONS

Recently initiated studies of aqueous plutonium chemistry are designed to provide information for modifications to the Purex flowsheet. There are some apparent advantages in operating the solvent extraction system at a low temperature, a low acid concentration, and without a reducing agent to partition plutonium. Neither the disproportionation nor the polymerization chemistry of plutonium is known for the specific flowsheet conditions, but studies are under way to obtain the relevant data.

The early results show that the low temperature and the presence of uranyl nitrate promote the Pu(IV) species in the aqueous system. Relative quantities of the III, IV, and VI states of plutonium have been determined for solutions with 0, 20, and 100 g/L of $\text{UO}_2(\text{NO}_3)_2$ at 5, 15, and 25°C. Plutonium distribution coefficients for these systems in equilibrium with the tributyl phosphate-*n*-dodecane (TBP-*n*DD) solvent will be determined.

11. SUPPORT TO ENVIRONMENTAL RESTORATION AND FACILITIES UPGRADE PROGRAM AND WASTE SOLIDIFICATION PROGRAM

The Environmental Restoration and Facilities Upgrade (ERFU) Program was initiated in early 1985 to facilitate ORNL's full compliance with all current environmental regulations and to plan for compliance with future regulations. The program provides comprehensive management for a large number of individual environmental restoration and improvement projects and activities, allowing ORNL to be fully responsive to its environmental protection needs. The Chemical Technology Division supports ERFU in four major areas: (1) coordination of long-range planning activities, (2) management of water pollution control projects for ORNL facilities at Y-12, (3) management of water pollution control projects at ORNL, and (4) strategy development and project management for air pollution control efforts.

In addition to ERFU support, Chem Tech is providing management for ORNL's Low-Level Liquid Waste (LLIW) Solidification Project. This effort will convert stored liquid waste with low-level radioactivity to solidified forms more suitable for long-term storage or disposal.

11.1 COORDINATION OF LONG-RANGE PLANNING ACTIVITIES

As part of the ERFU Program, systems for radioactive and hazardous waste collection, treatment, storage, and disposal are being improved or developed to provide adequate control over all ORNL-generated waste and effluents. Remedial actions are either under way or planned for all ORNL locations where past waste management practices or research activities have resulted in contamination of the facilities or the environment to levels that necessitate corrective actions.

To ensure effective management of these extensive environmental activities over the next several years, comprehensive long-range strategies have been developed that (1) outline the scope of needed improvements; (2) identify specific projects that

must be implemented; (3) determine the priorities of these projects based on regulatory compliance, health and safety concerns, schedule and budget constraints, and other programmatic considerations; and (4) develop overall cost and schedule estimates for use in budget requests to the Department of Energy (DOE). These strategies are continually updated to be consistent with current planning, and an annual ORNL Environmental Long-Range Plan is issued to summarize planning and strategy developments.

The *Preliminary ORNL Long-Range Environmental Management Plan* presents a program overview and summary of information on relevant regulatory requirements for each of the areas, overall strategy diagrams, and program costs and schedules. This information allows insight into the magnitude and direction of the environmental management program during the next several years. Chem Tech personnel provide coordination and integration of input from ERFU managers for all long-range planning activities.

11.2 WATER POLLUTION CONTROL PROJECTS FOR ORNL FACILITIES AT Y-12

Chem Tech also supplies management support for the water pollution control efforts for ORNL facilities at Y-12. This includes projects for the ORNL Biology Division facilities and the Isotopes Separation Facility to ensure that their discharges meet all requirements established by the Y-12 National Pollutant Discharge Elimination System (NPDES) Permit and the Y-12 Federal Facility Compliance Agreement (FFCA).

Biology Division Wastewater Cleanup

As required by the Y-12 FFCA, action must be taken to ensure that no Biology Division wastewater exceeding the limits established in the Y-12

NPDES Permit is discharged to East Fork Poplar Creek (EFPC). A new collection system and treatment facility are necessary to reduce the concentrations of specific pollutants in the Biology Division wastewater.

The treatment system will be designed to control pH and to remove suspended solids, trace organic compounds, and residual chlorine. It will also meet the "best available technology" (BAT) guidelines, comply with all effluent regulatory requirements, minimize sludge production, and minimize operation and maintenance requirements. Instrumentation systems with alarms will alert operators if the pollutant discharge limits are not being met or the facility is not operating normally.

Isotopes Separation Facility Wastewater Cleanup

The ORNL Isotopes Separation Facility is located in Building 9204-3 at the Y-12 Plant Site. Two recent capital projects have provided for new wastewater cleanup facilities at the building.

The first project funded alterations to allow the two existing sumps and pumps in the basement of Building 9204-3 to pump any spills to a slant-rib coalescing oil/water separator. The separated oil is then either reclaimed or packaged for shipment to a hazardous waste facility, while the oil-free water is discharged to EFPC.

The second capital project included funding for the pumps, piping, and tankage necessary for the collection of chemical wastewater. A tanker-truck loading station and ramp were also constructed to facilitate the transfer of chemical wastewater to the Y-12 wastewater treatment system.

11.3 WATER POLLUTION CONTROL PROJECTS AT ORNL

The diverse activities at ORNL generate a wide variety of liquid process wastes (PW), which, in combination with the natural characteristics of the site, present an extremely complex disposal challenge. Chem Tech provides management support for a number of the ORNL water pollution control projects.

The present PW system, shown schematically in Fig. 11.1, collects and processes liquid wastes that are normally not contaminated with radioactive materials but have the potential for contamination with varying types and quantities of chemicals. The PW system also collects water, including groundwater, that may be contaminated with very low levels of radioactivity, in addition to condensate from the low-level waste evaporator. Approximately 60% of this liquid waste is diverted to the Process Waste Treatment Plant (PWTP), where it is subjected to filtration and ion exchange before

Fig. 11.1. Current ORNL Process Waste System.

discharge to White Oak Creek (WOC). The remaining 40% is collected in PW ponds, monitored for radioactivity, and (if release criteria are met) discharged directly to WOC. The direct discharge of unmonitored and untreated process wastewater must be eliminated to comply with the ORNL NPDES Permit. To help meet this requirement, the Nonradiological Wastewater Treatment Project (NRWTP) will design and construct facilities to collect and provide appropriate treatment for all nonradiological PW at ORNL (see Fig. 11.2). In addition, a project is under way to provide on-line monitoring of trace metals in wastewater to allow optimization of the NRWTP waste treatment processes.

Nonradiological Wastewater Treatment Project

The NRWTP was begun in FY 1986, with a total estimated cost of \$18 million. Wastewater collection tankage (~6,800 m³) is to be provided at ORNL to replace seven active surface impoundments which must be removed from service by November 1988 to meet the Resource Conservation and Recovery Act (RCRA) requirements.

Pumping and piping will be installed to deliver collected wastewater to the Nonradiological Wastewater Treatment Facility (NRWTF) for treatment prior to discharge to WOC.

The new treatment facility will reduce contaminant concentrations to levels below the limits of BAT regulations established by the U.S. Environmental Protection Agency (EPA) under the Clean Water Act. Processes to be included in the NRWTF will remove suspended solids, trace metals, volatile organics, and semivolatile organics.

The proposed process flowsheet for the 2,700-m³/d (720,000-gal/d) treatment facility is shown in Fig. 11.3. Trace metals will be precipitated by chemical treatment with sodium hydroxide, followed by dual-media filtration to remove the suspended solids. Volatile organics are to be removed by air stripping, and final polishing is to be accomplished by passage through activated-carbon columns. The carbon columns will remove semivolatile organics and trace quantities of mercury. Caustic sludge produced by the process will be dewatered and packaged for disposal as a low-level radioactive solid waste.

Fig. 11.2. The ORNL Process Waste System as it will appear after completion of the Nonradiological Wastewater Treatment Project facilities.

Fig. 11.3. Proposed process flow diagram for the Nonradiological Wastewater Treatment Facility.

The established NRWTF flowsheet is based on wastewater characterization studies at ORNL and treatability studies done by Chem Tech.¹ The NRWTF schedule is controlled by the ORNL FFCA, and construction will be completed by September 1989.

Metals Monitoring

On-line metals monitoring systems are being installed at three locations at ORNL to provide real-time analysis for criteria trace metals (Cd, Cr, Cu, Ni, Pb, and Zn). The monitors will determine metal concentrations in wastewater streams, transmit real-time data, and provide signals for automatic diversion based on predetermined criteria. The real-time analysis and diversion system will increase the treatability of dilute streams contaminated with metals by eliminating dilution from streams not containing trace metals. The system will also reduce the volume of caustic sludge (low-level solid waste) produced at the NRWTF by diverting streams that do not require the metals removal treatment.

The metals analysis system to be installed at the NRWTF is based on one designed by Chem Tech personnel. The basic equipment, available commercially, includes an EG&G Princeton Applied

Research Corporation (PARC) Model 384B Polarographic Analyzer and two PARC Model 309 automatic voltametric electrodes. The system will incorporate a microcomputer to provide a continual flow of trace metals data to the ORNL Waste Operations Control Center and to automatically divert stream flow based on the preset diversion parameters.

11.4 AIR POLLUTION CONTROL

The objectives of the ERFU air pollution control efforts are to ensure that (1) air emissions from ORNL facilities are in compliance with applicable federal, state, and local regulations; (2) air emissions do not constitute a hazard to on-site personnel, the public, or the environment; and (3) emissions satisfy DOE's "as low as reasonably achievable" (ALARA) objectives. Chem Tech personnel on loan to the ERFU program provide support to these objectives through strategy development and management of projects designed to upgrade the air pollution control systems.

1. J. M. Begovich, C. H. Brown, Jr., J. F. Villiers-Fisher, and V. L. Fowler, *Treatability Studies in Support of the Non-radiological Wastewater Treatment Project*, ORNL/TM-10046, July 1986.

Air emission sources have been classified for the ERFU program as (1) radioactive air emission sources, (2) emission sources that require state permits, and (3) all other emission sources. Because much of the work done at ORNL facilities is related to nuclear energy, emissions containing radionuclides are of primary importance and have been given priority in the upgrade program. Emission sources that require state air permits are generally small and do not require extensive pollution-control equipment. Other emission sources that are not regulated by the Clean Air Act (CAA) have been identified but are not included in the ERFU Program.

Most of the air emissions containing radionuclides are discharged through seven major ventilation stacks. In addition, there are a number of exhausts from laboratory hoods and from individual vessels that vent locally through appropriate pollution-control equipment. Control of radioactive air emissions at the ORNL facilities is managed in accordance with DOE Orders 5480.1A, 5480.4, and 5820.2 and the CAA National Emission Standards for Hazardous Air Pollutants (NESHAP). Radioactive gas emissions from ORNL facilities are in compliance with existing regulations. Releases during 1985 resulted in a whole body dose commitment of 0.2 mrem (about 1% of NESHAP standards).

Because radioactive air emissions from ORNL facilities are in compliance with existing regulations, the program strategy is to identify and implement system upgrades needed to ensure continued regulatory compliance and to meet the DOE objectives for ALARA.

This strategy is being implemented through a series of technical studies and capital projects. The technical studies include a survey of ORNL stacks and vents to identify emission sources, inspections and evaluations to determine the condition of pollution-control equipment, and ALARA investigations to identify means to further reduce emissions in a cost-effective manner. Capital projects are in progress to upgrade several filter houses, repair underground ventilation ducts, and install emergency electrical generators for two off-gas and ventilation fans.

11.5 LIQUID LOW-LEVEL WASTE SOLIDIFICATION

Chem Tech is providing management support to the ORNL LLLW Solidification Project. This is a prototype effort to test the commercially available radioactive waste technologies for converting LLLW to solidified waste forms suitable for long-term storage and/or near-surface disposal. Liquid low-level wastes at ORNL are currently stored in eight 190-m³ (50,000-gal) tanks at the Melton Valley Storage Tank (MVST) Facility and in five tanks in Bethel Valley.

The LLLW Solidification Project is important for several reasons: (1) the LLLW storage capacity at the MVST Facility and in Bethel Valley is rapidly diminishing, and a shutdown of operating facilities at ORNL could occur if generation exceeds storage capacity; (2) the hydrofracture process is no longer authorized as a means for disposing of LLLW; (3) this project would process the LLLW stored on-site in inactive storage tanks which, under the provisions of the RCRA, ORNL could be ordered to remove at any time; and (4) without implementation of the LLLW Solidification Project, there is an increased chance that untreated waste could be released to the watershed with resultant potential risk to the public health.

Two LLLW solidification technologies, using cement and bitumen, have a substantial history of operation and will be the technologies used in the campaign to solidify 380 m³ (100,000 gal) of LLLW from the MVST Facility. The radioactive LLLW will be pretreated for decontamination, using filtration and zeolite beds. Following decontamination and solidification, the final waste form will be disposed of on-site by near-surface burial. This represents ORNL's long-term solution to the disposal of LLLW and is referred to as the "reference flowsheet" (Fig. 11.4).

Because of the rapid decrease in storage capacity at the MVST, the scope of this project also includes a short-term solidification campaign. Sampling analyses have shown that there is separation of the LLLW and the transuranic sludge inside tanks at the MVST, leading researchers to believe that the LLLW can be decanted from the upper

Fig. 11.4. ERFU reference process flowsheet for segregation, decontamination, and solidification of low-level liquid wastes at ORNL.

half of the tanks to produce a non-TRU solidified final waste form.² Therefore, in a near-term Emergency Avoidance Solidification Campaign (EASC), as much as 190 m³ (50,000 gal) of LLLW will be decanted from the upper half of the storage tanks and solidified to produce a non-TRU waste form. This flowsheet is shown schematically in Fig. 11.5.

The EASC will solidify the decanted LLLW in cement by processing 3.8 m³ (1,000 gal) of waste per treatment batch in a carbon steel liner. The liner will then be shielded in a concrete cask. After the cement has cured, the cement casks containing the waste and liner are to be transported by lowboy trailer to Solid Waste Storage Area 6 at ORNL for near-surface burial.

Ultrafiltration membrane studies using simulated LLLW are under way in Chem Tech, and in September 1987, a "pilot" study using actual LLLW to test zeolites and filtration membranes will be done in a hot cell facility at ORNL. The results of this study will help determine the final design of the decontamination and filtration columns for the reference flowsheet for LLLW solidification.

2. F. J. Peretz, B. R. Clark, C. B. Scott, and J. B. Berry, *Characterization of Low-Level Liquid Wastes at the Oak Ridge National Laboratory*, ORNL/TM-10218, December 1986.

Fig. 11.5. Process flowsheet for the ORNL Emergency Avoidance Solidification Campaign ("backup" flowsheet).

12. MISCELLANEOUS PROGRAMS

Because of its technical expertise and experience in many areas, Chem Tech has become involved in numerous small programs that are sponsored by DOE, other governmental agencies, or even private firms and organizations. Although these efforts do not represent a large fraction of our activities, they make important contributions to the sponsoring agencies. In addition, many are important to the national energy program and to the technical development and growth of the staff members involved. These efforts are viewed as essential parts of our overall mix of programs.

12.1 FUSION ENERGY STUDIES

During this report period, Chem Tech maintained its involvement in various aspects of the ORNL Fusion Energy Program. A conceptual flowsheet was prepared to illustrate the continuous processing of molten salt used as the blanket material for breeding both tritium and fissile material (^{235}U) in a fusion/fission hybrid reactor. The salt was 70 mol % LiF, 12 mol % BeF_2 , and 18 mol % ThF_4 . Preliminary cost estimates were prepared for the 3,000-MW(e) reactor.

In support of the pellet fueling program, Chem Tech designed and fabricated the gas handling system for the Joint European Tokamak pellet injector. A tritium pellet injector is also being designed and fabricated for use at the Tritium Systems Test Assembly at Los Alamos National Laboratory. In this device the pipe-gun concept is used for forming and pneumatically accelerating tritium pellets. Pellets are formed in situ in the breach by freezing tritium gas in a pellet-length cryostat. The pellets are then accelerated to the desired velocity with high-pressure gas. This device was chosen for tritium service because it has no moving parts inside the gun and because it requires no excess tritium to form the pellets.

12.2 THREE MILE ISLAND SUPPORT STUDIES

The involvement of Chem Tech and other ORNL staff members with recovery operations at the Three Mile Island Unit 2 (TMI-2) Nuclear Power Station has continued since shortly after the accident in 1979. In addition to the active participation of several Chem Tech staff members, the Division has managed all TMI-2 assistance provided by personnel at the DOE facilities in Oak Ridge, Tennessee.

Early assistance projects were coordinated through the Technical Advisory Group (TAG). This group was established in 1979 by General Public Utilities Corporation (GPU), the owner of the TMI-2 station. The four TAG members were experts from the national laboratories, and one was an ORNL staff member. The most significant TAG-coordinated project was the development by Chem Tech and Savannah River Laboratory personnel of the process used for (1) decontaminating the high-activity-level water (HALW) and (2) concentrating the radioactive contaminants into a readily disposable form.¹ The process was used to decontaminate $\sim 3,000 \text{ m}^3$ of HALW which had spilled into the reactor building basement and which remained in the Reactor Coolant System (RCS); these activities were completed in 1982. The process is still being used to clean up miscellaneous batches of water. A similar process is

1. E. D. Collins, D. O. Campbell, L. J. King, J. B. Knauer, and R. M. Wallace, "Development of the Flowsheet Used for Decontaminating High-Activity-Level Water," pp. 212-27 in *The Three Mile Island Accident: Diagnosis and Prognosis*, ed. L. M. Toth, A. P. Malinauskas, G. R. Eidam, and H. M. Burton, ACS Symposium Series 293, American Chemical Society, Washington, D.C., 1986.

being used in conjunction with defueling operations.

In December 1981, the Technical Assistance and Advisory Group (TAAG) was established by GPU to assist the TMI-2 operating utility, GPU Nuclear (a subsidiary of GPU), and its primary contractor for recovery operations, Bechtel National Corporation. The TAAG consisted of a variety of consultants from the nuclear power industry, the nuclear naval service industry, the Electric Power Research Institute, and the DOE national laboratories. Two of the seven members of the TAAG were Chem Tech personnel. Although the scope of advice and assistance provided by the TAAG was very broad, primary interests were on reactor disassembly, defueling operations, and facility decontamination. Perhaps the most important contribution was the suggestion and subsequent direction of video examinations of the reactor core, which led to the discovery of the large void area and the pile of rubblized fuel and core components. In February 1985, the TAAG sponsored another video examination. This time the subject of interest was the area beneath the reactor core. The examination showed the presence of a large mass of core materials (estimated to be ~20% of the total core) which appeared to have been melted during the accident. These discoveries were especially important for determining future defueling methods and equipment.

The TAAG was disbanded in early 1987, and, in its place, several ad hoc committees were formed. One of these had been formed in September 1986 to investigate problems that had occurred with maintaining clarity in the reactor vessel water during the defueling operations. Two of the 17 members of this committee are Chem Tech personnel. The committee has been responsible for the characterization of solid material within the water and for the application of coagulants, which have enabled longer filter life and improved maintenance of water clarity. Also, enhanced methods of video and ultrasonic imaging have been recommended.

Defueling operations at TMI-2 were begun in October 1985. Removal of the rubblized portion (~17%) of the reactor core was completed in early 1986. Water clarity during these operations was severely impeded by the formation of organic microorganisms (algae). This problem was studied thoroughly and finally solved by the periodic addition of H_2O_2 at a concentration level of 200 ppm.

Ten core samples were taken from the reactor vessel in July 1986 and sent to the Idaho National Engineering Laboratory (INEL) for use in a DOE-sponsored data acquisition program. The core-boring machine was then used to drill more than 400 holes into the once-molten region of the core. Removal of the created rubble is scheduled for early 1987. The intact fuel rods and assemblies on the bottom and periphery of the core, as well as the once-molten material (~20%) of the core which flowed into the bottom of the reactor vessel, remain to be removed.

Support projects recently carried out at ORNL included (1) a major project performed on short notice in September 1985 to conduct drop tests on one of the TMI-2 fuel canisters and to enable approval of the canisters by the U.S. Nuclear Regulatory Commission without a delay in startup of fuel removal operations; (2) the characterization of solid materials in the reactor vessel defueling water, as described above; and (3) studies of contaminated concrete core samples taken from the TMI-2 reactor building basement. The latter studies consisted of analyzing the results from leach tests made on the concrete samples at INEL and measuring pertinent properties of the concrete, such as density, porosity, and permeability. In addition, more extensive leach tests are planned at ORNL. All the preceding support projects were directed by Chem Tech personnel, although the work involved several other ORNL divisions.

12.3 RECOVERY OF URANIUM FROM DEGRADED PUREX SOLVENT

A number of solid sorbents were tested for their ability to remove uranium from the uranium-dibutyl phosphate (U-DBP) complex in 30% tributyl phosphate-normal paraffin hydrocarbon (TBP-NPH) solvents. A more complete description of this work is available.² In these tests, 30% TBP-NPH containing ~0.01 M each of uranium and DBP was used. In these studies, activated alumina was shown to remove both uranium and DBP from the solvents. Macroreticular cation-exchange resins or diethylenetriamine pentaacetic

2. J. C. Mailen and O. K. Tallent, "Recovery of Uranium from 30 vol % Tributyl Phosphate Solvents Containing Dibutyl Phosphate," pp. 381-95 in *Chemical Separations, Volume II: Applications*, ed. C. J. King and J. D. Navratil, Litarvan Literature, Denver, Colo., 1986.

acid (DTPA) on inert substrate removed the uranium and left the DBP in the solvent. Removal of valuable metal ions, such as enriched uranium, from the solvent before scrubbing with sodium carbonate solutions would prevent the metals from being stripped into the aqueous carbonate solution, from which their recovery is difficult.

Experimental Methods

The test columns were made of 8-mm-OD, 6-mm-ID glass tubing. A typical experimental setup is shown in Fig. 12.1. The solvent flowed down through the beds in all cases, and the flow rate through the packed beds was usually 1 mL/min. Uranium was determined spectrophotometrically in an aqueous ammonium carbonate strip solution using the absorbance at 450.4 nm, while DBP was determined by a procedure based on the quantity of uranium extracted by known concentrations of DBP.

Fig. 12.1. Apparatus used in small-bed tests.

Experimental Results

Macroreticular cation-exchange resin (Dowex 50W-X4, acid form, 50-100 mesh) was found to

load better at 52°C than at 22°C. Uranium loaded without detectable loading of DBP. Sample results are shown in Fig. 12.2. The uranium could be eluted either by TBP-NPH acidified by nitric acid or by aqueous nitric acid. The results of an elution test using 30% TBP-NPH containing 0.06 M HNO₃ are shown in Fig. 12.3. Higher acidity in the 30% TBP-NPH equilibrated with 2 M HNO₃ (0.36 M HNO₃ in the 30% TBP) resulted in faster elution, as shown in Fig. 12.4. Here, nearly all the uranium was eluted by 80 mL of acidified 30% TBP.

Fig. 12.2. Effluent uranium and DBP concentrations from a test loading of 1 mL of Dowex 50W-X4 cation-exchange resin (acid form) at 52°C. The flow rate of the solvent was 1 mL/min.

Fig. 12.3. Elution of uranium from 1 mL of Dowex 50W-X4 cation-exchange resin (acid form) by 30% TBP solvent containing 0.06 M HNO₃ at 22°C. The solvent flow rate was 1 mL/min.

Fig. 12.4. Duplicate tests of elution of uranium from 1 mL of Dowex 50W-X4 cation-exchange resin (acid form) at 22°C with 30% TBP solvent that had been equilibrated with an equal volume of 2 M HNO_3 . The flow rate of the solvent was 1 mL/min.

A column of 3-mm glass beads coated with ~0.2 g of DTPA (pH adjusted to ~7) was tested for removal of uranium and DBP from the solvent. After ~40 mL of effluent had been collected, the temperature of the column was increased from 22 to 50°C. At the lower temperature, the loading of uranium and DBP was minimal; at the higher temperature, the loading of uranium was significantly increased. This material appears to be usable for preferential removal of uranium from the U-DBP mixture. Removal of the loaded DTPA from the column is very simple since it is water soluble. Passage of a small amount of water (10 to 20 column volumes) through the column removed all the coating, with its uranium, from the column.

Possible Applications

The major potential use of the cation-exchange system would be to prevent the transfer of valuable metals (uranium and plutonium) to the solvent cleanup system, where they are more difficult to recover. These metals will be present in the solvent at a low level because of the strong complexes formed with DBP; low-acid stripping does not remove metals below about a 1:1 mol ratio to the DBP. Uranium and plutonium are the major metals present which form strong complexes with DBP; the plutonium complex is much stronger than the uranium complex.³ If the fuel being processed had a high uranium:plutonium ratio, as in

light-water reactor fuel, the quantity of plutonium held by the DBP would be quite small; however, in the case of breeder fuel the higher plutonium content would result in more retention of plutonium than uranium. The uranium and plutonium could possibly be removed from the solvent by a cation-exchange resin from which they could be easily recovered; the actual behavior of plutonium is not known.

Scrubbing with sodium carbonate solutions has been observed to produce emulsions, partly because of the presence of cations that form low-solubility complexes with carbonate; these cations include zirconium and plutonium.^{4,5} Treatment of the solvent with a cation-exchange resin to remove the cations that cause emulsions could significantly improve the operation of the solvent cleanup system. In fact, in the absence of cations, the solvent could be scrubbed with a basic solution without the necessity of a complexing anion such as carbonate. Weak bases such as hydroxylamine hydrate, which can be readily decomposed into innocuous gases with a significant reduction of solid wastes, would be attractive. The two applications discussed above assume that all cations can be removed from DBP solutions by cation-exchange resins. We have demonstrated the removal of uranium with the DBP remaining in the solvent. Macroreticular cation-exchange resins have been shown to remove zirconium and ruthenium from degraded solvent,⁶ but the cation removal rate, the capacity, and the fate of the DBP are not known. No studies of plutonium removal have been made. Applications such as those described above would require more testing with zirconium and plutonium.

3. L. Stieglitz, W. Ochsenfeld, and H. Schmieder, *The Influence of the Radiolysis of Tributyl Phosphate on the Plutonium Yield in the Purex Process for High Plutonium Content*, KfK 691, Karlsruhe Nuclear Research Center, 1968.

4. J. C. Mailen and O. K. Tallent, *Assessment of Solvent Cleanup Methods Using a Mixer-Settler System*, ORNL/TM-9118, 1984.

5. J. I. Kim, C. Lierse, and F. Baumgartner, "Complexation of the Plutonium(IV) Ion in Carbonate-Bicarbonate Solutions," pp. 318-34 in *Plutonium Chemistry*, ed. W. T. Carnall and G. R. Choppin, ACS Symposium Series 216, American Chemical Society, Washington, D.C., 1983.

6. C. N. Turcanu and D. Radu, "Sorption of ^{106}Ru and ^{95}Zr from Purex Solvent on Organic and Inorganic Ion Exchangers," *Radiochem. Radioanal. Lett.* 43, 245 (1980).

12.4 ASSESSMENT OF TRUEX DEPLOYMENT AT ROCKWELL HANFORD OPERATIONS

The solvent extraction process known as TRUEX (*TRAnsUranium EXtraction*) is being considered for use at the Rockwell Hanford Operations (RHO) Plutonium Finishing Plant (PFP). The purposes are to process the PFP waste, to recover the plutonium, to isolate the americium, and to convert the remaining waste to a nontransuranic waste. RHO requested that ORNL review the process and its possible deployment. This review⁷ addressed the generation of the TRUEX feed, the chemical flowsheet, and the products and raffinates. As a consequence of the review, it was suggested that present PFP operations be modified to reduce the concentrations of transuranium elements that will be in the TRUEX process feed. The review included an assessment of the TRUEX flowsheet on the basis of material balance, adequate extraction and stripping stages, and solvent cleanup. The final part of the review included results of three-party (RHO, ORNL, and Argonne National Laboratory) discussions of some major issues.

RHO provided funds for ORNL to evaluate the various processes for quantitatively converting the americium from the TRUEX process stream of low-concentration americium to a solid form suitable for future transfer to the Waste Isolation Pilot Plant. The evaluation recommended that removal of the americium onto an ion-exchange bed would be the best technology. The bed material could be fired for volume reduction, and the overall process would provide a volume reduction of 10^4 to 10^5 . After the evaluation, we began laboratory development to demonstrate the technology, and results of early experiments with europium support the 10^5 factor for volume reduction of the americium.

12.5 MOLTEN-SALT REACTOR DECOMMISSIONING

A technical evaluation of decommissioning planning for the former Molten-Salt Reactor Experiment, which was shut down in December 1969, has been completed. The preliminary results have been published,⁸ and a final report is in preparation.⁹ The key issues revolve around the treatment and disposal of ~5 metric tons of frozen fuel salt that contains >30 kg of fissionable ^{235}U plus fission products and higher actinides. The chemistry of

this material is complicated by the possible formation of elemental fluorine via a radiolysis reaction.

Supporting studies carried out as part of this evaluation include (1) an analysis of possible options, (2) calculation of nuclide decay in future years, (3) technical evaluation of the containment facility and hot cell penetrations, (4) review and update of surveillance and maintenance procedures, (5) measurements of facility groundwater radioactivity and sump pump operation, (6) laboratory studies of the radiolysis reaction, and (7) laboratory studies aimed at finding a suitable getter for elemental fluorine. In addition, geologic and hydrologic factors of the surrounding area were considered, as well as the implications of entombment in place with concrete.

The results of this evaluation show that the fuel salt cannot be left in its present form and location on a permanent basis. On the other hand, extended storage in its present form is quite acceptable for 20 or 30 years, or even longer. As a basis for future, permanent disposal, the material should be melted and repackaged into smaller containers to facilitate handling. During this operation, a fluorine getter can be easily added. The repackaged material can be stored for an additional period of time, if necessary, in a manner similar to remotely handled TRU waste, an operation already being conducted routinely at ORNL. Final disposal can then follow any number of pathways (none of which are actually available today but all of which are technically sound), including disposal in a high-level waste (HLW) repository, disposal in the Waste Isolation Pilot Plant, disposal via intermediate-depth disposal (also called greater confinement), or blending in with a fluoride-based HLW such as is processed at the Idaho Chemical Processing Plant for DOE. Utilization of the melt-repackage-dispose concept should save \$20 to \$50 million relative to the original reprocess-dispose concept. In addition, because melt-repackage is much simpler than reprocessing and uses only benign or inert chemicals, it poses a far smaller threat to both the environment and to the operators assigned to the task.

7. W. D. Bond, J. T. Bell, D. O. Campbell, and E. D. Collins, *ORNL Review of TRUEX Flowsheet for PFP Deployment*, ORNL/TM-10334, March 1987.

8. K. J. Notz, *Extended Storage-in-Place of MSRE Fuel Salt and Flush Salt*, ORNL/TM-9756, September 1985.

9. K. J. Notz, *Decommissioning of the MSRE—A Technical Evaluation*, ORNL-6379, to be published.

During the course of this study, an excellent getter for elemental fluorine in a radiation field was discovered. A patent disclosure is being filed on this process.

12.6 RECOVERY OF ALUMINA AND OTHER USEFUL MINERALS FROM FLY ASH

The economics of coal ash utilization appears to depend on the ability to utilize large fractions of the ash, not just to recover a few relatively high-value products such as alumina. The most recent stage of active research on recovery of useful materials such as alumina from coal ash was completed; further work in the area depends upon assessments of the results. Initial assessments of the concepts made by Kaiser Engineers, the major participant in the Electric Power Research Institute program, were most favorable. The quantities of residual solids (principally, mullite and silica for ashes from most eastern U.S. coals) are so large that utilization of these materials can greatly affect the overall economics of ash processing. Kaiser Engineers, with assistance by the Ontario Research Foundation, has found potential uses for the residue; and some uses for limited quantities of the residue could bring relatively high prices. Of course, the high-value products would require additional separations; and their markets are limited. A particularly attractive use is as filler material for industrial plastics, and Ontario Research was able to make physical separations to produce relatively good materials from our leached residues.

The most recent ORNL work involved a survey of potential ash sources and characterization of the trace impurities in different phases of ashes. Twenty-two ash samples from different utilities were analyzed and characterized. Although the ashes differed significantly in composition, all but two have favorable economic potential for processing, under the criteria set up by Kaiser Engineers. The two exceptions were ashes from western coals, which contained too much calcium and thus required too much acid to make processing attractive. These assessments depended strongly upon the criteria set up by Kaiser Engineers, particularly the market value and market size for several of the products.

Additionally, the relatively large number of well-characterized ash samples available made it possible to gain greater insight into the behavior of many elements in coal ash. With dissolution data from all the ashes and with reliable chemical

analyses of ash, leach solutions, and leached residue, the distribution of several elements between the soluble and insoluble phases was available. The principal phases of interest in ashes from eastern coals are believed to be the insoluble mullite phase, the soluble amorphous silicate phase, and the largely soluble iron oxide phase(s). By comparing the solubility of an element with that of aluminum, which distributes between the mullite and the amorphous silicate phase, and with iron, which is largely in the iron-oxide-based phases, one can estimate the distribution of several elements. Although the differences in the ashes introduce considerable scatter in the results, several clear and interesting trends were apparent. For instance, the alkali metals and even other alkaline earth elements such as magnesium are not as leachable as calcium and thus tend to have higher distributions into the insoluble phases than calcium.

12.7 EMERGENCY RESPONSE STUDIES FOR THE FEDERAL EMERGENCY MANAGEMENT ADMINISTRATION

As a portion of a comprehensive effort by the Federal Emergency Management Administration (FEMA) to evaluate the strengths and weaknesses of the nation's preparedness for emergencies that might arise from hostile acts, accidents, or natural causes, two studies were initiated within Chem Tech. In both cases the goals were to evaluate the threat and to suggest means of diminishing exposure to disaster. The topic of the first program is the expedient production of antibiotics. The second program is concerned with the problems associated with the release of airborne hazardous materials.

Expedient Production of Antibiotics

In a catastrophic disaster that would destroy capabilities for producing antibiotics and at the same time create conditions under which epidemic diseases and large numbers of injuries would occur, the availability of broad-spectrum antibiotics could be of major importance to the survival and recovery of a major portion of the threatened population. The purpose of this project is to identify processes that would allow expedient production of antibiotics under austere conditions such as would follow a nuclear attack.

Five antimicrobial drugs—penicillin, cephalosporin, tetracycline, bacitracin (topical), and sulfonamide (chemically produced)—were identified

for emergency production. Antibiotic manufacturing plants in the continental United States, Mexico, and Puerto Rico were identified, along with potential alternative sites for expedient antibiotic production, such as producers of single-cell protein, enzymes, and fermentation ethanol.

This investigation revealed that a typical antibiotic manufacturing facility is composed of two main sections: (1) a highly specialized and relatively small fermentation unit and (2) a very large (80 to 90% of the total plant area) separation and purification unit. The fermentation section requires specialized equipment, which is not readily available elsewhere, for operation in a sterile environment. The emergency production of antibiotics under austere conditions will be feasible only if a substantial reduction in the separation and purification sections of the plant can be realized. Other specific results of the study are given elsewhere.¹⁰

Countermeasures to Hazardous Chemical Releases

Recent major incidents involving the release of hazardous chemicals have heightened the awareness of both the public and private sectors that effective strategies must be developed to prevent and deal with emergencies. Although the whole area of hazardous materials may require such a treatment, the most chronic needs are for strong direction in cases where an airborne hazard is involved. The materials might be in any form, but the means by which the threat overtakes people is such that little warning is possible and immediate means of protection are very limited. A number of federal, state, and local government agencies share portions of the responsibility for various aspects of the problem. With the very considerable overlap in responsibility as well as gaps in the coverage, it was considered desirable to perform a study of the entire collage of activities and to recommend appropriate roles for FEMA and other agencies.

A comprehensive study of FEMA's role required that the project be divided into a number of tasks. To date, the first of four tasks has been completed. In reviewing the major airborne disasters of the past decade, it was evident that no single factor stood out as the predominant cause of these chemical releases. Therefore, it was deemed necessary to assess the level of threat to which the country is exposed by the potentially most dangerous chemicals. The methodologies available prior to the

current study were deemed inadequate, and therefore a new method was developed as a major contribution to the first task.

The factors contributing to hazard were individually studied. For instance, it can be shown that the annual number of releases of chemical species is directly proportional to their annual production rate. The major factors that influence hazard are toxicity level, fire or explosive hazard, mobility of the substance after release, production rate, and plant location and domestic shipments. A correlation was developed to allow the prediction of relative threat, τ_r , as a function of the contributing factors. This correlation is given as follows:

$$\tau_r = (N_H \cdot M_H + N_F \cdot M_F + N_R \cdot M_R) \cdot [N_M \cdot M_M \cdot (N_P \cdot M_P + N_S \cdot M_S)] ,$$

where N is the rating, M is the multiplier, and the subscripts are defined as follows:

- H = toxicity,
- F = fire,
- R = explosion,
- M = mobility,
- P = production,
- S = shipments.

Fifteen chemicals were selected with which to test the correlation. Their selection was expected to give a good cross section of the chemical industry, from large volume to the specialty product with high toxicity. The ranking of chemicals based on the Environmental Protection Agency's Acute Hazardous Events data base of injuries and/or deaths from 1980 to 1985 followed the prediction very well indeed for a very large body of information. It was concluded that the recommended correlation be expanded to a larger data base for further testing. This work is in progress.

10. P. R. Bienkowski, D. D. Lee, and C. H. Byers, "Evaluation of Separation and Purification Processes in the Antibiotic Industry," presented at the 9th Symposium on Biotechnology for Fuels and Chemicals, Boulder Colo., May 5-8, 1987; to be published as a special issue of *Applied Biochemistry and Biotechnology*.

12.8 INDUSTRIAL CONSERVATION— MULTICOMPONENT SEPARATIONS BY CONTINUOUS ANNULAR CHROMATOGRAPHY

The central objectives of this work are to accelerate the industrial development of the continuous annular chromatograph (CAC) concept and to maintain a research program specifically aimed at solving outstanding problems in implementation while broadening the acceptance of the concept.

The CAC was developed in the Chem Tech Division over the past decade. The development allows the performance of chromatographic separations in a truly continuous and steady-state fashion. A bed of granular sorbent material such as ion-exchange resin is contained in the annular space between two concentric cylinders. A stationary port is used to feed the mixture that requires separation to the top of the bed. The bed itself slowly rotates past the feed port. Eluent is continually distributed over the top of the bed. The separated products are collected at exit tubes equally spaced around the periphery at the bottom of the rotating bed.

The CAC is one of the very few unit operations that are capable of multicomponent separations. It is inherently capable of any separations that can be obtained through conventional liquid chromatography. Some limitations, such as dispersion in the bed and the dilution effect, which is characteristic of adsorptive chromatography, must be met to improve the efficiency of the process and hence broaden its utility as an industrial unit operation.

The first exploratory implementation of the CAC in the biotechnology area was for the separation of fructose-glucose-sucrose mixtures, which was investigated experimentally in a laboratory-scale CAC using calcium-exchanged Dowex 50W-X8 resin as adsorbent.

Figure 12.5 shows that complete resolution of fructose-glucose mixtures could be obtained both in a 60-cm-long CAC and in a conventional column of the same length with sugar feed concentrations up to 200 g/L. Partial resolution of sucrose in three component mixtures was also obtained, and the three sugars were completely separated from higher-molecular-weight saccharides added to the feed mixtures as impurities.

The experimental results were analyzed using orthogonal collocation methods in which linearized chromatographic parameters were used for both fixed and rotating beds. Bed properties and

Fig. 12.5. Comparison between experimental and calculated concentration profiles for CAC operation at base conditions.

equilibrium and mass transfer parameters used in the model were obtained through independent experiments. A typical orthogonal collocation result for a single component is given in Fig. 12.6.

ASPEN, a unique process and economic simulator, is used to obtain a valid comparative evaluation of a conventional separation technique with the new, innovative, continuous chromatographic method. ASPEN simulates any process, performing both mass and energy analyses as well as economic analyses. Since annular chromatography is a new technique, a new module was written in FORTRAN to simulate the CAC. This process has been completed, and the continuous chromatography module has been tested in a conceptual butanol fermentation process (Fig. 12.7). Using this process simulator as a comparative device for different approaches to separation offers absolute commonality of basis. With these capabilities, we will be able to use, adjust, and modify the codes as needed to analyze both the chromatographic and conventional approaches to several types of separations. In addition to the process simulator module for the continuous chromatograph, work is proceeding on a cost-estimating model for the CAC unit. The analyses are focused on energy consumption in the process alternatives, but overall economics will also have to be considered to ensure

Fig. 12.6. Comparison of the analytical and orthogonal collocation solutions for standard CAC conditions.

Fig. 12.7. Butanol fermentation/separation—ASPEN simulation flowsheet.

that there will be sufficient overall justification for adopting a new process technique. Except when capital costs become excessive, energy consumption (including energy invested in reagents) and process costs are usually closely related. An industrial liaison group has been established to aid in the selection of appropriate processes for comparison. Their experience and data are used wherever possible to enhance both the importance and the accuracy of the results.

12.9 IMPROVED COAL LIQUEFACTION BY USE OF CONTROLLED-TEMPERATURE CONTACT WITH SOLVENTS

A small experimental effort was made to explore a concept first developed at ORNL in 1982. The yield of useful liquids from direct (solvent) coal liquefaction was found to depend upon the temperature history of the coal solvent during heatup to reaction conditions, not just upon the rate of heatup. The preliminary experiments had indicated that, with Kentucky No. 9 coal and a Lummus solvent, the yield of liquid products could be increased from 60 to 70% to almost 90% by holding the coal-solvent slurry at an intermediate temperature for a few minutes before continuing the heating to reaction conditions (at ~ 700 K).

Recent experiments confirmed the earlier results and identified the optimum intermediate temperature as ~ 589 K. The time required at the intermediate temperature was not determined, but it appears to be less than 5 min. The generality of the approach was investigated by examining a different coal (Illinois No. 6) and a different solvent (a recycle solvent from the Wilsonville, Alabama, coal liquefaction pilot plant). Neither the new coal nor the new solvent appeared to give yields as high as those observed in the first experiments with Kentucky No. 9 coal and Lummus solvent, either with linear heatup rates or with selected programmed heating procedures. However, in all cases, the programmed heating that involved holding the slurry at an intermediate temperature for a few minutes improved the liquid yield significantly, usually $\sim 30\%$. Since the experiments with the different coal and/or different solvent were more limited, it is possible that optimum conditions were not evaluated. Future studies may show higher yields with these materials.

A preliminary evaluation of the quality of the resulting liquids was made by determining the compositions of oils, asphaltenes, and preasphaltenes. In all cases tested, the fraction of oils in the liquids was higher and the fraction of preasphaltenes was lower in the liquids produced by programmed heating. This implies that the resulting increased quantity of liquid product may also require less hydrogen for upgrading.

The data available thus far are not sufficient to prove a mechanism for the enhanced liquid yields or even to prove that upgrade of the liquids will be less costly in hydrogen consumption. However, we have proposed a mechanism for the effect and are investigating the proposed mechanism.

It appears likely that the optimum intermediate temperature corresponds to a temperature at which the coal becomes more porous and probably swells slightly but undergoes little reaction. Heating Kentucky No. 9 coal to the optimum intermediate temperature with solvent and cooling (without heating further to the reaction temperatures) shows no visible change in the coal particles but probably some solvent uptake.

If solvent is allowed to penetrate the coal particles more completely at this intermediate temperature before the coal is heated to reaction conditions, the solvent, by being in more intimate contact with the coal, could be far more effective in transporting hydrogen from hydrogen-rich centers to hydrogen-poor centers, even within individual coal particles. This should make the solvent considerably more effective in liquefaction. The time required at the intermediate temperature is that necessary for the solvent to diffuse into and penetrate the coal particles effectively.

12.10 VAPOR-LIQUID EQUILIBRIA OF FLUORIDES IN UF_6

There is an economic incentive to use vapor-phase samples of UF_6 for determining the concentration of impurities in cylinders of UF_6 rather than using liquid-phase samples, as has been done in the past. Calculation of liquid-phase concentrations from vapor-phase sample analyses would rely on a thermodynamic vapor-liquid equilibrium model. The potential of such an approach has been shown previously.¹¹ A vapor-liquid equilibrium model for UF_6 and some of its common impurities would have other useful applications in the industry for purification, enrichment, and conversion of UF_6 for nuclear power plant fuel.

A vapor-liquid equilibrium model for UF_6 and its common impurities has been implemented on the computer; a data bank of component parameters has been developed for use with the model; and the results of preliminary calculations have been reported.¹² The model incorporates the Haydon-O'Connell equation of state to represent nonidealities of the multicomponent vapor-phase

mixture. The UNIQUAC model is used for liquid-phase activity coefficients.

Hydrogen fluoride, an important impurity in UF_6 , exhibits the unusual property of forming association complexes (including a cyclic hexamer) in the vapor phase. Such behavior is extremely difficult to model. An empirical model of HF - UF_6 vapor-liquid equilibrium has been found to be the most practical approach to modeling this system.¹³

A program has been initiated to acquire equilibrium vapor and liquid samples from cylinders of UF_6 at the Paducah Gaseous Diffusion Plant. These samples will be analyzed for concentration of the common impurities, and the results will provide a data set from which to regress UNIQUAC binary interaction parameters and to demonstrate the capabilities of the vapor-liquid equilibrium model.

12.11 CHEM TECH DIVISION FACILITIES

The Chem Tech Division occupies facilities that are scattered throughout the X-10 and K-25 sites. Table 12.1 summarizes the current space utilization by Chem Tech in these areas. Over the past 18 months, we have undertaken and accomplished a major cleanup of a very broad spectrum of buildings and facilities. After cleanup and/or renovation by Chem Tech, several buildings have been transferred to other ORNL divisions or returned to productive use by our own staff members. These are discussed in the following section.

General Upgrade of Buildings

Building 2528. In Building 2528, a cleanup effort was required to remove and package forty-three 55-gal drums of organic-contaminated carcinogenic material. After repurmitting is received for this

11. W. Davis, Jr., *Liquid-Phase Compositions from Vapor-Phase Analyses: I. Thermodynamic Properties of UF_6 and Some of Its Potential Impurities*, K/OA-5578, Oak Ridge Gaseous Diffusion Plant, 1983.

12. H. D. Cochran and W. Davis, Jr., *Liquid-Phase Compositions from Vapor-Phase Analyses: II. Adaptation of the Prausnitz Program to UF_6 and Some of Its Potential Impurities*, K/OA-5762, Oak Ridge Gaseous Diffusion Plant, 1985.

13. W. Davis, Jr., and H. D. Cochran, *Liquid-Phase Compositions from Vapor-Phase Analyses: III. Reanalysis of HV Vapor Densities and of UF_6 -HF Liquid-Vapor Equilibria*, K/OA-5904, Oak Ridge Gaseous Diffusion Plant, 1986.

Table 12.1. Space utilization by Chem Tech Division

Building	Total area ^a (ft ²)	Number of occupants ^b	Function
K-1006	2,254	8	Research, offices
K-1037	4,923	10	Offices
2528	1,874	0	Standby (high bay)
3001	1,385	10	Offices
3017	5,242	10	Offices, computer laboratories
3019	7,070	24	Offices, hot cells, research, storage
3503	3,527	8	Research, offices
3541	800	1	Research
3592	1,200	3	Research
45G-N	21,272	88	Research, offices
4501	21,395	34	Research, offices, hot cells
4505	9,579	26	Research, offices
4507	2,655	0	Standby (hot cells)
7920	5,369	26	Research, hot cells, offices
7930	1,907	9	Research, hot cells, offices
Total	90,452	257	

^aIncludes research areas, offices, utility rooms, conference rooms, rest rooms, computer laboratories, hot cells, basement and attic space, and change houses.

^bIncludes personnel from Chem Tech and other supporting divisions.

building, it will house the Chem Tech dust-handling facility and waste research.

Building 3013. After this geologic disposal laboratory was renovated using General Plant Project (GPP) funds, it was released to the Operations Division for safe storage of the environmental monitoring computer tapes.

Building 3019. Throughout its more than 40 years of service, Building 3019 has had many additions and modifications. Since many of these changes were made to accommodate small programs, there has rarely been adequate funding to permit upgrading of the building. As a result, most of the major systems (e.g., off-gas, liquid waste, ventilation, emergency, and containment) are conglomerates remaining from the various activities. In response to DOE's request to place the building in standby and to use it only for storage of solid, fissile materials, we have actively undertaken significant efforts to eliminate those deficiencies in the building systems which would preclude safe standby. A new waste line has been installed in the west end of the building. Environmental Restoration and Facilities Upgrade (ERFU) funding has been obtained to clean up the contaminated hot cells in the west end of the building and to replace cracked, leaking windows in the hot

cells. GPP funds are being used to replace the emergency evacuation system. The refurbishing efforts to place this facility in a safe standby condition will continue through 1990.

Building 3503. Building 3503 had become a storage area for Chem Tech and other divisions to house unwanted furniture, equipment, and materials. After a substantial cleanup effort, this facility was used in the following successful research endeavors:

1. studies to assess the impact of adding mop water from the Y-12 Plant to the K-25 sanitary sewage wastewater treatment plant,
2. equipment and process development for the continuous production of uranium gel spheres,
3. development of equipment and procedures for the removal of hydrofluoric acid from gas streams with solid sorbents,
4. pilot studies of a wet-air oxidation system to treat wastewaters contaminated with high concentrations of organic compounds, and
5. development and testing of equipment used in Building 3019 for the production of (U,Pu)O_x

microspheres [Plutonium-Uranium Oxide Microsphere Preparation (PUMP) Program].

From the storage garden east of Building 3503, 17 contaminated radioisotope casks (many belonging to the Operations Division) were properly wrapped and sealed in plastic and buried in an approved manner. (Several of these casks had become unwrapped and were emitting 30 mrem of radioactivity per hour.)

Southwest of Building 3503, a concrete pad was cleaned, and twenty-three 55-gal, rusty drums (some leaking) containing solvents, thorium, and mixed wastes were disposed of safely. Core samples taken from under the pad revealed contamination. The concrete pad area had been submitted for decontamination and decommissioning efforts under the ERFU umbrella; the pad will be removed during the next fiscal year.

Building 3508. In Building 3508, a very extensive and comprehensive decontamination program was carried out which included the following:

1. extensive repairs to the heating, ventilating, and air-conditioning unit;
2. cleaning and removing 26 highly contaminated glove boxes;
3. cleaning and removing a massive lead-shielded carrier contaminated with transuranium elements;
4. removing and decontaminating a highly contaminated cave liner and support equipment; and
5. cataloging and disposing of large amounts of diverse chemicals.

For the first time, the entire laboratory area in this building was decontaminated to the point that protective clothing was no longer required for entry. Upon completion of the cleanup activities (at Chem Tech expense), the building was released to the Instrumentation and Controls Division.

Building 3541. This process development laboratory, which was previously vacant, was decontaminated and then reclassified from a C-zone to a regulated zone. This building is currently being used as an active research facility by the Environmental Control Technology Group of the Engineering Development Section of Chem Tech.

Building 4501. In the basement of Building 4501 was a neglected and forgotten room housing an unsecured radiological hazard in the form of con-

taminated ion-exchange columns that contained low-level contamination (^{237}Np and plutonium). Loose, contaminated equipment was safely disposed of; existing ion-exchange columns were drained; and the room was decontaminated and sealed.

In addition, the basement storage area and stairwells of this building were inventoried, and the following disposition of the contents was made: (1) thirteen 4- by 8-ft copper screen sections were taken to K-25 for surplus sale; (2) five skid loads of fittings, flanges, valves, pumps, and instruments were smeared and salvaged; and (3) 60 lb of abandoned asbestos insulation was transferred to Y-12 for burial.

Laboratories BG-73 and BG-74 were cleaned up and converted to engineering facilities, with only the hoods being classified as C-zones.

In Room 125, 9 of the 15 wall vaults used for storage of intermediate-level radioactive materials and 50 of the 59 below-floor storage wells used for storage of high-level radioactive materials were emptied and decontaminated. In accordance with approved safety procedures, 185 liquid samples were discarded to the high-level liquid waste system.

Building 4507. Two deteriorated flammable-liquid storage cabinets (containing 55 bottles of liquid organic chemicals) were physically located by the southeast entrance to Building 4507. Wind currents were continually causing an accumulation of tree leaves, cups, and papers close to the storage cabinets. This potential fire hazard was removed by properly inventorying the chemicals and requesting the Environmental and Occupational Safety Division's assistance in disposing of them. The cabinets were ultimately salvaged.

Staff Relocations to K-25 Site

In order to establish a multisite presence for the Waste Management Technology Center (WMTC), Chem Tech negotiated with Oak Ridge Gaseous Diffusion Plant (ORGDP) management for space at the K-25 site to house the WMTC. Agreements were reached which allowed the WMTC staff to physically locate in the east end of Building K-1037. The building was modified to allow representatives of private sector companies to meet with WMTC staff within the ORGDP complex.

Demonstrations of waste management technologies by private companies may be held at a variety

of locations on the Oak Ridge Reservation. Since Building K-1423 at ORGDP is located outside the security fence and is equipped with offices, change rooms, and high-bay areas with utility services, it has been identified as a focal point for conducting many of these demonstrations.

Another group in the Chem Tech Division, Engineered Waste Disposal Technology, has also been relocated to the ORGDP site. This group of approximately ten employees resides in Building K-1006 and provides support to Chem Tech's Waste Management Analysis Research and Development staff and to the WMTC.

12.12 SUPPORT TO ENERGY SYSTEMS ENGINEERING

During this report period, Chem Tech continued to work closely with Martin Marietta Energy Systems, Inc., Engineering on several projects.

Technical Support to the FMPC

Energy Systems Engineering is assisting the Feed Materials Production Center (FMPC), Fernald, Ohio, in several projects for upgrading the production capability of the plant. One project is the Low-Level Waste Processing and Shipping System. Chem Tech personnel, in cooperation with the Analytical Chemistry plus the Systems and the Equipment Technology departments of ORGDP, have been assisting Energy Systems Engineering in this effort. This effort consisted of characterizing the various wastes requiring treatment for shipping, estimating both the backlog and steady-state quantities to be treated, preparing the Conceptual

Design Report with assistance from Lockwood Greene, sampling and characterizing the kiln off-gas, and proposing flowsheets. However, because of over-budget estimates, the project is being reevaluated.

Chem Tech personnel are providing technical support to Energy Systems Engineering in designing and modifying the plant for UF_6 to UF_4 reduction at FMPC. Other FMPC projects for which Chem Tech personnel have provided assistance consist of the following: decladding of reject materials; NO_x scrubbing; upgrading of the solvent extraction system; and upgrading of the general, Plant 8, and refinery sumps. The Analytical Chemistry and Equipment Technology departments of ORGDP participated in these sump projects.

Technical Support to Y-12 Projects

Chem Tech is also providing technical support to Energy Systems for various Y-12 projects. One project involves the renovation and upgrading of the existing chemical recovery equipment to meet production needs. Another project is the survey of the existing effluent stacks for environmental improvement project planning.

Chem Tech is providing technical support to Energy Systems Engineering in the S-3 Pond closure. The Tennessee Department of Health and Environment has approved the closure plan for the S-3 Ponds. This closure will consist of in situ grouting of the biodegradation sludges, covering the grout with clay, capping the area with a double liner that contains a drain fabric between the liners, and adding a final cap for sod.

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AWARDS, HONORS, AND RECOGNITIONS

J. M. Begovich

1985 Publications Award, Martin Marietta Energy Systems, Inc.

1985 International Technical Communication Award of Distinction in Scholarly and Professional Articles Category, Society for Technical Communication

J. T. Bell

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1985 Chairman, Fourth International Symposium on Separation Science and Technology for Energy Applications

1986 Chairman, Expert Panel on Instrumentation, International Federation of Clinical Chemistry

1986 National Lectureship Award, American Association for Clinical Chemistry

W. D. Bond

1985 IR-100 Award for development of an improved metal oxide varistor, *Industrial Research*

1986 Invited by the National Research Council to serve as a member of the Advisory Panel for the Center for Analytical Chemistry, the National Bureau of Standards

W. D. Box

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

D. O. Campbell

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

D. T. Brooksbank

1986 Technical Communication Award of Excellence in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

S. D. Clinton

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

C. A. Burtis

1984-1986 Board of Directors, American Association of Clinical Chemists

H. D. Cochran, Jr.

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1985 Technical Communication Award of Distinction in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

E. D. Collins

1985 Achievement Award for Safeguards Upgrade of Building 3019

J. L. Collins

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

S. R. Dinmore

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

K. E. Dodson

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

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- 1986 Technical Communication Award of Excellence in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

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- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

W. R. Gambill

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

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- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1985

First Annual M. Sargis (Sarge) Ozker Award for contributions in radioactive waste management, American Society of Mechanical Engineers

1986

Chairman, Standards Working Group 16.6, standard leach test for low-level radioactive waste, American Nuclear Society

E. Greenbaum

1985

Technical Communication Award of Merit in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

1985

Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1985

Significant Event Award for the development of metallized chloroplasts, Martin Marietta Energy Systems, Inc.

1986

International Technical Communication Award of Achievement in the Scholarly and Professional Articles Category, Society for Technical Communication

1986

Fellow, American Association for the Advancement of Science

P. A. Haas

1985

Golden Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1985

Inventor Award, Martin Marietta Energy Systems, Inc.

D. W. Holladay

1985

Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

A. R. Irvine

1985

Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

E. K. Johnson

- 1986 Special Recognition Award for elimination of radiological, environmental, and safety hazards in Chemical Technology Division facilities, Martin Marietta Energy Systems, Inc.

R. L. Jolley

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.
- 1986 Distinguished Service Award, Division of Environmental Chemistry, from the American Chemical Society
- 1986 Program Chairman, Division of Environmental Chemistry, American Chemical Society
- 1985 Technical Communication Award of Excellence in Books Category, East Tennessee Chapter of the Society for Technical Communication
- 1986 International Technical Communication Award of Excellence in Books Category, Society for Technical Communication

D. S. Joy

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

A. D. Kelmers

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

J. M. Kennerly

- 1983-1986 Public Relations and Publications Chairman, Newsletter Editor, Knoxville-Oak Ridge Section of the American Institute of Chemical Engineers
- 1986-1987 Secretary, Knoxville-Oak Ridge Section of the American Institute of Chemical Engineers

N. E. Lee

- 1985 Technical Communication Award of Distinction in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication
- 1985 International Technical Communication Award of Distinction in Scholarly and Professional Articles Category, Society for Technical Communication
- 1986 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

R. E. Leuze

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

B. A. Lewis

- 1986 Margaret Oakley Dayhoff Award, Biophysical Society

T. B. Lindemer

- 1985 Publications Award, Martin Marietta Energy Systems, Inc.
- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.
- 1985-1988 Trustee, Nuclear Division, American Ceramic Society

M. H. Lloyd

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

R. S. Lowrie

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

J. C. Mailen

- 1985 Golden Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

E. W. McDaniel

- 1985 Technical Communication Award of Excellence in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication
- 1985 Session Chairman, Conference on Hydrofracture and Grout Techniques, jointly sponsored by the U.S. Department of Energy and the University of Arizona
- 1985 International Technical Communication Award of Achievement in Scholarly and Professional Articles Category, Society for Technical Communication

C. P. McGinnis

- 1985 Chairman, Knoxville-Oak Ridge Section, American Institute of Chemical Engineers
- 1985 Achievement Award for CEUSP Operation, Martin Marietta Energy Systems, Inc.
- 1985 Newsletter Editor, Nuclear Engineering Division, American Institute of Chemical Engineers
- 1986 Operational Performance Award, Martin Marietta Energy Systems, Inc.
- 1986 Newsletter Editor, Nuclear Engineering Division, American Institute of Chemical Engineers
- 1986 Engineer of the Year, Knoxville-Oak Ridge Chapter, American Institute of Chemical Engineers

D. R. McTaggart

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

J. R. Mroczek

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.
- 1986 Inventor's Award, Martin Marietta Energy Systems, Inc.

K. J. Notz, Jr.

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

I. W. Osborne-Lee

- 1986 Vice President, East Tennessee Chapter of the National Organization for the Professional Advancement of Black Chemists and Chemical Engineers

W. L. Pattison

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

R. L. Pearson

- 1985 Publications Award, Martin Marietta Energy Systems, Inc.

W. H. Pechin

- 1986 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

W. W. Pitt, Jr.

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

D. J. Pruett

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

B. R. Rodgers

- 1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.
- 1986 Chairman and Keynote Speaker for the Environment at the Joint Chinese/American Institute of Chemical Engineers Symposium, Taipei, Taiwan
- 1986 Chairman for the Environment, World Congress of Chemical Engineering III, Tokyo, Japan

1985-1987 Director, Fuels and Petrochemicals Division, American Institute of Chemical Engineers

1986-1988 Chairman of Program Committee, Fuels and Petrochemicals Division, American Institute of Chemical Engineers

R. G. Ross

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

R. Salmon

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

F. M. Scheitlin

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

R. M. Schultz

1985 Award of Excellence, Nuclear Weapons Program, Y-12, Martin Marietta Energy Systems, Inc.

C. D. Scott

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1986 Member, National Academy of Engineering

1986 Inventor of the Year Award, Martin Marietta Energy Systems, Inc.

M. B. Sears

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

S. E. Shell

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

D. E. Shepherd

1986 Certified Professional Secretary

W. E. Shockley

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

S. P. N. Singh

1984-1990 National Laboratories Consortium Representative to the Emissions and Control Technologies Task Group (Task Group I), National Acid Precipitation Assessment Program

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

M. G. Stewart

1985 Technical Communication Award of Distinction in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

1985 Technical Communication Award of Merit in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

1986 Technical Communication Award of Excellence in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

1986 International Technical Communication Award of Achievement in Scholarly and Professional Articles Category, Society for Technical Communication

G. W. Strandberg

1986 Appointed Adjunct Associate Professor, Department of Microbiology, University of Tennessee, Knoxville

O. K. Tallent

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

L. M. Toth

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

V. C. A. Vaughan

1982-1987 Cochairman, Ethics Committee, Knoxville-Oak Ridge Section of the American Nuclear Society

J. F. Villiers-Fisher

1985 Golden Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

J. S. Watson

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

D. J. Weaver

1985 Certified Professional Secretary

R. M. Wham

1985 Secretary, Oak Ridge-Knoxville Section, American Institute of Chemical Engineers

1986 Treasurer, Oak Ridge-Knoxville Section, American Institute of Chemical Engineers

S. K. Whatley

1985-1986 National President, Society of Women Engineers

1986 Governor, American Association of Engineering Societies

J. Woodward

1985 Technical Communication Award of Merit in Scholarly and Professional Articles Category, East Tennessee Chapter of the Society for Technical Communication

1985 Significant Event Award for development of a simple and inexpensive glucose assay based upon the coupling of glucose oxidase and Fenton's Reagent, Martin Marietta Energy Systems, Inc.

R. G. Wymer

1985 Silver Acorn Award for patent development, Martin Marietta Energy Systems, Inc.

1986 Technical Communication Award of Excellence in Periodic Activity Reports Category, East Tennessee Chapter of the Society for Technical Communication

1986 Fellow, American Institute of Chemical Engineers

CONSULTANTS

J. T. Bartis

Affiliation: EOS Technologies, Inc.
Expertise: Fossil Energy Technology
Number of Days: 8
Section/Program: Engineering Coordination and Analysis

J. R. Benemann

Affiliation: Georgia Institute of Technology
Expertise: Applied Biology
Number of Days: 3
Section/Program: Basic Energy Sciences

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Affiliation: University of Washington
Expertise: Chemical Engineering
Number of Days: 2
Section/Program: Chemical Sciences

A. L. Boch

Affiliation: Independent
Expertise: Management
Number of Days: 10
Section/Program: Pilot Plant

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Affiliation: University of New Mexico
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Number of Days: 3
Section/Program: Sedimentary Rock

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Affiliation: Independent
Expertise: Bioprocess Engineering
Number of Days: 6
Section/Program: Biotechnology

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Number of Days: 5
Section/Program: Chemical Development

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Section/Program: Energy Research

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Section/Program: Chemical Development

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Expertise: Chemistry—Portland Cement Hydration and Expanding Cements
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Section/Program: Engineering Development

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Affiliation: Duke University
Expertise: Genetics—Chloroplast DNA in *Chlamydomonas*
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Affiliation: CSA, Inc.
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Section/Program: Engineering Development

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Section/Program: Energy Research

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 Section/Program: Nuclear and Chemical Waste

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 and Development

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 Section/Program: Chemical Sciences

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 Expertise: Thermodynamics
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 Section/Program: Chemical Development

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 Expertise: Thermodynamics—Advances in Multi-
 Component Phase Equilibria
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 Section/Program: Chemical Development

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 Expertise: ANFLOW Technology—Anaerobic
 Treatment
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 Section/Program: Engineering Development

C. Sola

Affiliation: Universitat Autònoma Barcelona
 Expertise: Chemical Engineering
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 Section/Program: Energy Research

F. Toldra

Affiliation: Institute de Agraquímica de Tecnologia de Aliments
 Expertise: Treatment of Waste and Wastewater
 Using Anaerobic Fluidized-Bed
 Reactors
 Number of Days: 1
 Section/Program: Energy Research

H. B. Ward

Affiliation: University of Mississippi
 Expertise: Microbial Coal Liquefaction
 Number of Days: 10
 Section/Program: Energy Research

SEMINARS

- July 15, 1985 **"Microbial Processes for Petroleum Recovery,"**
T. F. Yen, University of Southern California, Los Angeles, California
- January 30, 1986 **"Biotechnology and the Future,"**
A. E. Humphrey, Lehigh University, Bethlehem, Pennsylvania
- July 17, 1986 **"Process Engineering in a Changing Industry,"**
J. R. Fair, University of Texas, Austin, Texas
- September 11, 1986 **"Development of an Automated, High-Pressure, Heat Flux**
Calorimeter for Natural Gas Hydrate Studies,"
J. S. Lievois, Rice University, Houston, Texas
- September 16-18 **"Facility for the Analysis of Chemical Thermodynamics,"**
(FACT Short Course)
C. W. Bale and A. D. Pelton, Ecole Polytechnique de Montreal,
Montreal, Quebec, Canada
- October 10, 1986 **"Chemical Engineering Activities at the National Bureau**
of Standards,"
J. Hord, Center for Chemical Engineering, National Bureau of Standards,
Boulder, Colorado
- October 15, 1986 **"Adaptive Optimization of Continuous Bioreactors,"**
H. C. Lim, Purdue University, West Lafayette, Indiana
- October 23, 1986 **"Molecular Thermodynamics for Chemical Process Design,"**
J. M. Prausnitz, University of California, Berkeley, California
- November 17, 1986 **"The Ligninolytic System of *Phanerochaete chrysosporium*:**
Role of Molecular Oxygen and Relationship to Secondary Metabolism,"
B. D. Faison, Procter and Gamble Company, Cincinnati, Ohio
- November 24, 1986 **"Digital Systems for the Control of Fermentation and**
Separation Units,"
J. A. Phillips, Lehigh University, Bethlehem, Pennsylvania
- December 9, 1986 **"Expert Systems Software Development for Assessment**
of Solid Waste Leaching and Disposal,"
T. Constable, Wastewater Technology Centre, Environment Canada,
Burlington, Ontario, Canada

INTERNAL PROGRAM REVIEWS

DATE	PROGRAM	COORDINATOR
May 7, 1985	Miscellaneous Programs (Seed Money, New Initiatives, etc.)	E. K. Johnson
September 10, 1985	TRU/TRUST	R. E. Leuze
October 15, 1985	Sedimentary Rock	A. G. Croff
November 19, 1985	NRC—Waste	S. K. Whatley
December 3, 1985	NRC—Safety	R. A. Lorenz
January 14, 1986	Waste Technology Development Programs	J. A. Klein
February 11, 1986	Interactions with ORNL Nuclear and Hazardous Waste Programs	R. K. Genung
March 18, 1985	Radioactive Waste (IDB and Repository)	J. A. Klein
April 15, 1986	Biotechnology	T. L. Donaldson
May 20, 1986	Consolidated Fuel Reprocessing Program	J. T. Bell
June 17, 1986	Engineering Support (ORNL, K-25, Y-12, Waste Assistance)	V. C. A. Vaughen
July 15, 1986	Chemical and Material Sciences	J. S. Watson
August 13, 1986	Reactor Programs	R. P. Wichner

**CHEMICAL TECHNOLOGY DIVISION STAFFING LEVEL AND FINANCIAL SUMMARY
FY 1986**

	Funding (in thousands)	Chem Tech Scientific/Technical ^a (person-years)	Total Scientific/Technical ^b (person-years)
DOE PROGRAMS			
Coal	\$422	2.8	2.8
Gas	3		
Petroleum	0		
Breeder Reactor Systems	1,617	10.9	12.9
Remedial Action	165	0.8	0.8
Nuclear Fuel Cycle	30		
Defense Waste and By-Products Management	2,754	13.8	16.8
Nuclear Waste Fund	4,535	14.8	17.0
Transportation	97	0.9	0.9
Multisector	179	1.3	1.3
Materials Production	5,474	25.5	29.0
Environmental R&D	414	2.6	2.6
Basic Energy Sciences	7,664	39.7	43.3
Energy Research Analysis	45	0.1	0.1
Environmental Compliance	379	1.7	2.7
SUBTOTAL	\$23,778	114.9	130.2
DOE-RELATED PROGRAMS			
Nuclear Regulatory Commission	\$2,608	13.5	14.1
Other DOE and Contractors	1,913	6.9	7.2
DOE-ORO Contractors	446	3.1	3.8
State	6		
Other Energy Systems Plants	247	2.1	2.1
SUBTOTAL DOE-RELATED PROGRAMS	\$5,220	25.6	27.2
TOTAL DOE PROGRAMS	\$28,998	140.5	157.4
WORK FOR OTHERS			
State and Local Government	247	1.4	1.4
Other Federal (Excluding NRC)	251	1.4	1.5
Other Department of Defense	140	1.1	1.1
TOTAL WORK FOR OTHERS	638	3.9	4.0
MULTISITE SUPPORT			
Hazardous Waste	30		
TOTAL	\$29,666	144.4	161.4

^aChem Tech research staff supported by the Division in FY 1986.

^bTotal research staff supported by Chem Tech funding in FY 1986, including personnel on loan to Chem Tech from other divisions.

[illegible]

INTERCOMMITTEE RESEARCH
 C D SCOTT¹
 INTERMEDIATE AND PHYSICAL
 RESEARCH
 E CHEMISTRY
 F LEE
 J WOODWARD
 O A GRAVES²
 P SUBSIST
 C V FEVANS
 M WATTS¹⁰
 G C WATKINS¹⁰
 INTERMEDIATE RESEARCH
 C D SCOTT¹
 D B DAVIDSON
 D B LEE
 I B LEWIS
 J E FERGUSON
 K A WOODWARD
 L A CRYST³

CHEMISTRY RESEARCH
 1. 0.00000000
 RESEARCH ON LONG-RANGE SOURCE
 1. 0.00000000
 1. 0.00000000
 RESEARCH AND MEASUREMENTS
 RESEARCH
 C. A. BUDNICK
 B. F. GAGAN

W L CARRINGTON
L S BENTLEY
S M BENTLEY
J L WELLS

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TRANSPORTATION TECHNOLOGIES
1. B. SHAPIRO
D. S. MOY
P. C. ARNONE

[illegible]

ENVIRONMENTAL CONTROL TECHNOLOGY
C H SHOWN JR
J M SGOVICH
R M COUGHS
O W SHAFER
V L FOWLER
J M SGOVICH
E W FISHER
E WILSON

J L HEWITT
M L JENNINGS
D R HOFFGARD

GROUP TECHNOLOGY DEVELOPMENT
E W McDermid
D B DeLozier
T S Adams
D A Tallent
G D DeLozier
R E Goodson
C L Francis
T F Goodsey
D B Trotter

(44) HENRIETTA WATTS BISHOP 1922-1981
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 A V WELFORD
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 S C OSBORNE

SPECIAL ASSIGNMENTS
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F A HAAS
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D W STANLEY
DIP SAFETY OFFICER
D HALL

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AFFIRMATIVE ACTION
 REPRESENTATIVE
 C. E. LADD

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D R LINDENBROOK

SAFETY, RADIATION CONTROL, AND
ENVIRONMENTAL PROTECTION
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**QUALITY ASSURANCE COORDINATOR
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42. J. L. Collins	85. D. W. Holladay
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87. T. D. Hylton
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- 90–114. E. K. Johnson
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- 131–145. C. E. Lamb
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- 198–202. C. D. Scott
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Document Reference Section
244. S. K. Whatley	281-282. Laboratory Records Department
245. J. H. Wilson	283. Laboratory Records, RC
246. K. Wilson	284. ORNL Patent Section
247. J. Woodward	

EXTERNAL DISTRIBUTION

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- 297-298. R. M. Bernero, Deputy Director, Office of Nuclear Material Safety and Safeguards, US Nuclear Regulatory Commission, Washington, DC 20555
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