The High-Level Waste Immobilization Program: An Overview

W. F. Bonner

September 1979

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INTRODUCTION

Research and development of nuclear waste immobilization has been under way at Pacific Northwest Laboratory (PNL) since the 1950s. Currently, the Department of Energy (DOE) is sponsoring a major technology development program at PNL for treating and handling high-level nuclear wastes (HLW) in preparation for final disposal. This paper describes some major aspects of that program.

High-level waste immobilization research began at PNL mainly to prepare technology for treating waste produced by commercial fuel reprocessing. This early program, designated the Waste Solidification Engineering Prototypes Program, was terminated at PNL in 1970. In 1972 the program now entitled the High-Level Waste Immobilization Program (HLWIP) was initiated. The objective of the HLWIP is to provide the technology for converting radioactive waste to nondispersible forms suitable for handling, transportation and disposal. Although initial emphasis was on commercial waste, it was determined in 1976 that this technology is also applicable to wastes stored at various U.S. sites. Since 1977 the program's emphasis has been on high-level defense wastes.

Several waste forms were evaluated during the Waste Solidification Engineering Prototypes Program. The first form studied was calcine; examination of phosphate glass, phosphate-ceramics, and borosilicate glass followed. As a result of this program, borosilicate glass was later selected as the most promising waste form for further development.

Although the current program has emphasized borosilicate glass development, it has also established and maintained a strong program for developing alternatives to the monolithic glass waste form. In 1973 PNL established contracts with Pennsylvania State University for development of a crystalline waste form, later called "supercalcine." Shortly thereafter, PNL initiated development of coating technology for pelletized waste forms and began studying a multiple-barrier waste form. Study of alternative waste forms will assure that concepts with specific advantages for second-generation plants are not overlooked and that all waste forms are being evaluated on an equal basis. Many waste forms are being investigated at PNL in an effort to identify the best possible forms for full-scale use. Forms being considered include concrete, glass beads, supercalcines, glass-ceramics, cermets, and SYNROC.^(a)

Technology developed in this program can be applied to solidification of high-level radioactive wastes now in storage at Savannah River, Hanford, Idaho National Engineering Laboratory and West Valley, New York. High-level wastes resulting from commercial power production and other wastes such as transuranium-contaminated wastes and low-level wastes can also be treated using this technology.

(a) SYNROC is an acronym for synthetic rock--a crystalline waste form developed at the Australian National University in which waste components are incorporated into selected mineral phases.

The technology development pattern for the program is illustrated in Figure 1. First, radioactive wastes are characterized and laboratory-scale development of suitable waste forms is conducted. These waste forms are then evaluated. If found to be suitable, the waste forms are subjected to engineering studies that include pilot-scale process development. If the waste form and process are suitable, large-scale demonstrations are conducted. Full-scale canisters of solidified waste are then characterized to verify that the process can reliably produce waste forms of appropriate quality.

FIGURE 1. High-Level Waste Immobilization Program Technology Development Pattern

HIGH-LEVEL WASTE

High-level waste was first generated in large quantities during World War II as part of the U.S. effort to develop the atomic bomb. Commercial nuclear power plants generate spent fuel, which can be reprocessed to recover uranium and plutonium for recycle. Spent fuel reprocessing also results in production of liquid HLW.

High-level liquid waste is the aqueous stream resulting from the first solvent-extraction step during fuel reprocessing. It has the following characteristics:

- it contains over 99% of the radionuclides produced in the nuclear fuel cycle;
- it is a nitric-acid slurry as generated, but is neutralized with sodium hydroxide at many sites before storage in tanks;
- levels of heat produced by radioactive decay in liquid HLW make it necessary to provide cooling systems that prevent self-boiling;
- radiation protection requires that all processing be performed behind thick concrete walls.

Numerous fuel reprocessing flowsheets have given rise to many HLW chemical compositions. Wastes stored at each site (Hanford, Savannah River, Idaho Engineering Development Laboratory, and West Valley, NY) are different, and compositions vary markedly from tank to tank within each site. Composition ranges for both defense and commercial HLW are shown in Table 1. Because of the many waste composition variations, HLW forms and immobilization processes must be quite insensitive to waste changes.

TABLE 1. Composition Ranges in Representative High-Level Wastes

Constituent	Commercial, wt%	Defense, wt%
$\text{Na}_2\text{O}/\text{Na}_2\text{CO}_3$	0-10	0-10
Fe_2O_3	3-29	6-61
Cr_2O_3	1-2	0-27
NiO	1	1-11
Al_2O_3	0	0-86
MnO_2	0	4-40
TiO_2	0	0-18
Gd_2O_3	0-31	0
CaF_2	0-5	0-54
Na_2SO_4	0	0-5
U_3O_8	0-12	2-14
Fission-Product Oxides	39-88	1
Actinides	5-15	1

The radioactivity and heat-generation rate of HLW decreases with time. Table 2 shows heat generation and radiation from a canister of solidified commercial HLW. A canister 1 ft in dia and 10 ft tall contains solidified HLW from 2.5 tonnes of spent nuclear fuel. It is worth noting that after 1000 yr the canister's heat-generation level matches that of a very small light bulb. After this 1000-yr storage period a person would have to stand within one foot of an unprotected canister for over two minutes to receive a radiation dose equivalent to that of a standard chest x-ray.

Unlike many other toxic substances such as lead or mercury, the toxicity of radioactive waste steadily decreases. Figure 2 (Heckman 1977) compares the toxicity of several materials as a function of time. Toxicity index in this figure is determined by the amount of water required to dilute the materials shown to levels considered safe for drinking water. As shown in Figure 2, the toxicity of a HLW repository is lower than that of common lead and mercury ore bodies after approximately 500 years. Although waste forms are designed to remain intact much longer than 500 yr, one may conclude that precautions beyond those employed with ore bodies may not be cost-effective after this period.

High-level waste resulting from commercial nuclear power production represents a relatively small volume. Should U.S. policy allow reprocessing of spent nuclear fuel, the resulting HLW volume from a typical reactor would approximate that shown in Figure 3. If all the electrical power for a city the size of Seattle (750,000 people) were generated by a nuclear reactor (only one reactor required), about ten canisters of solidified HLW would be produced in one year if glass were selected as the waste form. Projecting an increasing individual power demand--by the year 2000, the 338 waste canisters produced powering this city of 750,000 people could be stacked on 10% of a single tennis court. If these canisters were buried in an underground repository, the space required would be less than the area beneath one mile of a four-lane highway.

TABLE 2. Heat Generation and Dose Rates from a Typical HLW Canister as a Function of Time

<u>Decay Time, yr</u>	<u>Heat- Generation Rate, Watts</u>	<u>Dose Rate 1 ft From Surface, R/h</u>
1	22,000	110,000,000
10	3,100	62,000
100	360	5,800
1,000	20	1.6
10,000	6	1.3
100,000	3	0.6

FIGURE 2. Toxicity Index of Spent Fuel and High-Level Waste

FIGURE 3. Commercial High-Level Waste Volume

(a) In actual storage or disposal the canisters would be spread apart for cooling purposes.

WASTE FORM DEVELOPMENT

Once liquid waste characteristics are determined, waste forms can be developed to immobilize the radionuclides therein. In selecting a waste form, resistance to radionuclide release is a prime consideration. The form should, under both normal and accident conditions, be resistant to dispersion by water-leaching, air currents, and vaporization. Since all waste form production must be done remotely--with operators shielded by concrete walls--the equipment involved must be unusually simple. The radioactive waste form selection process requires the use of scientists and engineers experienced in both waste form development and remote processing.

Because of its inertness, its ability to accommodate waste composition variations, and its relative ease of production, glass has received the greatest development effort internationally during the past few decades. Nevertheless, promising crystalline and non-crystalline waste forms have also been identified, as have barriers such as inert-material coatings. An increasing amount of attention is being focused upon the merits of these alternative waste forms. Forms and barriers currently of interest are described below.

GLASS

Research on drying and vitrifying (glassifying) HLW has been under way since the 1950s. From this effort, a standard concept of a large canister filled with borosilicate glass has evolved. Formulations have been developed for the major quantities of HLW stored in the U.S., as well as for wastes that may be produced from processing of spent commercial nuclear fuel.

Literally thousands of glass formulations have been tested to zero in on quality glass compositions. Drawing upon this information pool, a generic study of waste form compositions vs. glass properties has been initiated. This comprehensive study will require two years to complete, but will encompass all major planned HLW glass compositions in the country. Upon completion of this study, glass property changes resulting from changes in waste glass components will be predictable.

ALTERNATIVE WASTE FORMS

Although glass has shown outstanding inertness and ease in processing over a broad range of conditions, other waste forms offer even greater inertness in certain environments. For example, under high-temperature and high-pressure conditions some crystalline phases resist dissolution by water substantially better than does glass. A number of alternatives to the glass waste form are consequently being investigated.

Universities and other organizations offering specialized knowledge have been contracted to assist PNL in this area. One such organization (Pennsylvania State University) has developed the crystalline waste form "supercalcine" in this program. By adding certain chemicals to the liquid waste, specific crystals containing radionuclides may be formed during drying and subsequent heat treatment. These crystals may be tailored to tightly hold radioactive elements in a given geological environment.

Non-crystalline forms such as glass marbles and partially crystallized forms such as glass-ceramics are also being developed and analyzed.

BARRIERS

If radioactive wastes are encased in an impermeable container, hazardous radionuclides obviously cannot escape. Studies under way with various waste form coatings and container designs indicate that virtually total containment for a few hundred, and perhaps thousands of years is not only possible but is probably achievable with a modest effort.

As discussed previously, after 500 yr HLW toxicity is comparable to that of ore bodies found throughout the world. Barriers that will contain the waste form during this period are being developed. One such concept for protecting the entire waste canister is shown in Figure 4. This concept provides: 1) engineered containment through modification of nearby geologic media; 2) highly inert overpacks of carbon, ceramic, polymer, metal or other material; 3) a metallic canister; and 4) a quality waste form such as glass or crystalline material.

FIGURE 4. Engineered-Barrier System

Further waste form inertness is provided on a smaller scale through the multibarrier concept shown in Figure 5. In this concept, waste forms are shaped into pellets or beads up to 1 cm in diameter. These pellets or beads are coated with inert, nonporous materials such as carbon or ceramics, then encapsulated in a matrix inside a canister. The matrix material provides adequate thermal conductivity as well as improved containment and strength.

FIGURE 5. Multibarrier Product

Many multibarrier waste forms have been fabricated and tested at PNL. Figure 6 shows cut and polished one-kilogram samples of four waste forms. Coated waste forms such as these have been under development in this program for several years. Coatings and barriers offer the potential for total waste form isolation from the surroundings for hundreds and perhaps thousands of years.

FIGURE 6. Multibarrier Product Samples

WASTE FORM CHARACTERIZATION

The waste form is a primary barrier for containing radionuclides; therefore, knowledge of waste form characteristics is essential. This program includes a major effort for characterizing the stability of glass and alternative waste forms. Properties such as leachability at various temperatures in different solutions, susceptibility to radiation damage, effects of high-temperature storage, friability, potential for volatilization, density, homogeneity and others are being evaluated. These tests are performed in laboratories with both simulated and actual radioactive waste forms.

An alternative waste form characterization and comparison study is under way which includes major waste forms under development in the U.S. and abroad. Testing and comparison of these forms on a uniform basis provides information on the relative merits of each.

LEACHABILITY

The possibility of groundwater coming into contact with HLW in an underground repository represents a potential radionuclide-release situation. Radionuclides dissolved in groundwater may eventually reach man's environment. Consequently, an important aspect of HLWIP work is the determination of waste form resistance to water-leaching. Tests include variations of groundwater composition and pH, temperature, pressure, and leachant-volume-to-surface-area ratios. Typical data from this program are shown in Figures 7 and 8.

Leaching tests conducted under this program have shown that both glass and crystalline waste forms may be altered in water at high temperatures. Waste repositories are designed with the expectation that they will remain dry for hundreds of years. However, should water penetrate a repository, it is desirable that the repository temperatures not be significantly in excess of 100°C at the time the water actually contacts the waste container. At such temperatures the chance of substantial radionuclide release into groundwater is remote.

FIGURE 7. Comparative Chemical Durabilities

FIGURE 8. Release of Radionuclides from Glass During Leaching

RADIATION/HEAT EFFECTS

Particles emitted by radioactive decay can break chemical bonds in the waste form, thereby decreasing its resistance to dispersion mechanisms. Tests to determine the effects of radiation are generally conducted on an accelerated basis: the waste forms (glass and other potential materials) are "doped" with radiation-emitting isotopes. Waste glass samples prepared in this manner, such as the one shown in Figure 9, have simulated the effects that over a half million years of radiation exposure would have upon a typical commercial waste glass. These samples have maintained their integrity without significant changes.

FIGURE 9. "500,000-Year-Old" Waste Glass

Radiation damage experiments with crystalline waste forms show that some crystals are resistant to radiation, while others are destroyed. This knowledge makes it possible to develop crystalline waste forms that exhibit improved radiation stability. Further tests are being conducted on several waste forms to determine what the effects are when radioactive atoms in the matrix transmute to different elements during radioactive decay.

Self-heating in the waste glass, which results from the decay of radioactive elements, has the potential effect of raising the glass temperature to a point where crystallization (known as "devitrification") may occur. To test the effects of devitrification, samples of the waste-containing glasses are heated to selected temperatures and held at that point for up

to five years. These samples are then leach-tested for long periods to determine changes in chemical durability. X-ray diffraction and scanning-electron-microscope evaluations are also performed on the heat-treated samples to establish the type, size and distribution of the crystals. Tests show that properties relating to potential radionuclide release from the waste glass change only slightly as a result of devitrification.

IMPACT BEHAVIOR

Radionuclides could accidentally be released into the atmosphere if a radioactive waste form were broken into small enough pieces and if protective barriers failed. Impact characteristics of glass-filled canisters are therefore evaluated to determine the potential for dispersion during accident conditions. Included in these studies is a series of tests in which a regular production canister filled with solidified glass is dropped on its bottom corner onto an unyielding surface. Canisters such as those shown in Figure 10 have suffered only minor damage; internal glass fracturing was localized. After these drop tests the canisters are opened and the contents are evaluated for the quantity and size of fine particles produced. These determinations make it possible to calculate the increase in glass surface area. Such tests have shown a canister of waste glass to be impressively tough.

FIGURE 10. Drop-Test Canister

PROCESS DEVELOPMENT

Several systems for solidifying radioactive wastes are being evaluated and developed as part of the HLWIP at PNL. Major efforts have been concentrated upon the following:

- the heated-wall spray calciner
- the fluidized-bed calciner
- the joule-heated continuous melter
- the in-can melter
- the disc-pelletizer
- the marble-maker.

Two calciners and two melters have been developed to provide backup technology. Alternative waste form equipment installed or planned for assessment at PNL includes presses, sintering equipment and metal-matrix equipment. These are being evaluated and modified to improve their ability to function under totally remote conditions.

Since nonradioactive testing and development is many times more efficient than is equivalent in-cell work, most process development work is conducted using simulated high-level wastes. Nonradioactive simulated wastes are chemically the same as the anticipated radioactive wastes, except that nonradioactive isotopes are used and some minor substitutions are made.

DRYING AND CALCINATION

Most currently conceived waste immobilization programs require drying of radioactive liquids and conversion of the resulting solid to an oxide form. This process is referred to as "calcination." At PNL both the heated-wall spray calciner and the fluidized-bed calciner have been developed for use with HLW. The end result in either system is a fine, dry powder consisting of inert solids and radioactive elements.

Calcination of liquid wastes is normally considered to be only the first step in the immobilization of nuclear wastes. Conversion of this powder to a larger waste form, such as a glass or a crystalline form, substantially reduces its susceptibility to dispersion by blowing air or flowing water and increases its ability to disperse heat generated by radioactive decay.

Heated-Wall Spray Calciner

Figure 11 shows the important features in the spray-calcination process--a leading system for calcining radioactive waste. Liquid waste is sprayed into the heated chamber where liquid components are quickly evaporated and drawn off through filters, allowing the calcined powder to fall into the chamber below. The calciner shown in Figure 12 is the largest of three spray calciners at PNL. Operating at demonstrated feedrates in excess of 400 g/h, just one of these systems would be large enough to effectively process the liquid HLW currently stored at Savannah River in less than 15 years. If spent fuel were reprocessed, a system this size could dry and calcine HLW at a high enough rate to service more than 100 nuclear power reactors--enough electrical power for 75 million people.

FIGURE 11. Spray Calciner

The spray-calcination systems at PNL have successfully logged thousands of operating hours in radioactive and nonradioactive operations. This proven system is being redesigned for inclusion in the planned Savannah River Waste Immobilization Facility.

FIGURE 12. Plant-Scale Spray Calciner/Continuous Melter

Fluidized-Bed Calciner

The fluidized-bed calciner is a backup calcination system for HLW processing. The unit developed by PNL is shown in Figure 13. This system is easier to operate and more flexible than conventional fluidized-bed calciners. During operation, a continuous stream of silica particles is added to the fluidizing chamber; combustion of kerosene and oxygen in the bed provides the necessary heat. Liquid waste is sprayed into the hot bed, where liquids quickly flash off as steam. The resulting calcine coats the silica particles and overflows into the waste-treatment process or receiving chamber below.

FIGURE 13. Fluidized-Bed Calciner

VITRIFICATION

One well developed waste immobilization process combines calcined solids with glass-forming chemicals, then melts the resulting "batch" to form a glassy, vitrified solid. Depending upon the melting method and subsequent treatment steps used, the resulting glass form may range in size from a small marble up to a large, canister-filling monolith. Some glass-filled canisters are shown in Figure 14.

Currently, two vitrification systems are being developed at PNL for use in waste management: joule-heated continuous melting and in-can melting.

FIGURE 14. Canisters of Simulated High-Level Waste Glass

Joule-Heated Continuous Melting

The mainline process being prepared for immobilization of HLW stored at the Savannah River Plant is the coupled spray calciner and continuous melter shown schematically in Figure 15. The continuous-electric-melter system has been adapted from melters in operation in the glass industry. The melter is simply a refractory-lined cavity with electrodes at opposite ends. Molten glass between the electrodes is heated by dissipation of electric current in the melt. The melter is enclosed in a stainless-steel shell to contain radioactive vapors and to allow their discharge to an appropriate treatment system.

FIGURE 15. Spray Calciner/Ceramic Melter System

During operation, calcined HLW and glass-forming frit from the spray calciner enter the melter and form a cool layer on the molten glass. The calcine and frit slowly melt into the glass pool. Molten glass from the pool flows up a riser and overflows into the receiving canister below. Melt-flow termination to allow filled-canister replacement is accomplished by tilting the melter two to four degrees. A more recent melt-draining technique includes air-lifting the melt from the melter, thus avoiding the need to tilt the melter.

The continuous melter is attractive because of its high capacity, long life, and excellent glass quality. The spray calciner/continuous melter shown in Figure 12 has been operated for over 1000 h, producing glass at rates often exceeding two tonnes per day.

It is possible to run a very high level of power through the continuous melter's electrodes, thus rendering the melter capable of direct vitrification of liquid waste. Since a calciner is not required in this system, this type of operation significantly reduces the overall system complexity. The melter shown in Figure 16 was designed for direct liquid feeding. It has converted over 10,000 ℓ of liquid waste to glass at rates of over 100 ℓ /h.

FIGURE 16. Joule-Heated Continuous Melter for Liquid Feeding

In-Can Melting

The in-can melter is a well developed backup vitrification system. In this system, diagrammed in Figure 17, the canister itself is used as the melting crucible. Heat for melting is provided by a cylindrical furnace surrounding the canister. During in-can melter operation the melter is coupled to a calciner, from which it receives calcine and frit and to which it discharges effluents. Calcine and frit flow into a 1050°C canister, where the melting

FIGURE 17. Spray Calciner/In-Can Melter Process

occurs. Internal metallic fins can be used to increase heat transfer and melting rate significantly. After a canister is filled, the calcine-frit stream is diverted to an adjacent melter. Adequate time is thus available for completing all melting steps and replacing the filled canister.

The spray calciner/in-can melter equipment shown in Figure 18 became operational in April 1978. Since that time, hundreds of operating hours have been logged at glass production rates often exceeding 100 kg/h. This system's relative simplicity, advanced state of development, demonstrated high melting rates, ability to handle troublesome compositions, and advantage of essentially replacing the melter with each new canister make it attractive as a radioactive waste immobilization process.

FIGURE 18. Plant-Scale Spray Calciner/In-Can Melter

ALTERNATIVE WASTE FORM PROCESS

Decisions regarding the desirability of alternative waste forms must include an evaluation of the system by which the form is made. Since HLW immobilization equipment will operate in a production mode within concrete containment walls and without hands-on maintenance, only simple systems with a few moving parts are practical. Some processes may be prohibitively costly and unreliable when operated on a fully remote basis.

This program includes evaluation of current technology for producing alternative waste forms and provides comparisons on a uniform basis. Process selections for further development are based upon these evaluations. Drawing upon the experience of remote-operation designers and pilot-plant engineers, equipment is prepared and installed in PNL's pilot-plant facility, where it can be tested as a complete system.

One waste form being tested on an engineering scale involves placement of glass beads in a metal matrix. A system for manufacturing the glass beads, shown in Figure 19, has been developed and is undergoing large-scale tests. Molten glass draining from a continuous melter flows into molds on an endless chain. The vibrating molds cool and shape the molten glass into beads. Beads drop out of the molds into a canister below. The continuous melter/bead-former system shown can produce over 50 kg of beads per hour.

FIGURE 19. Glass Bead Forming Machine

Production of small beads formed from glass produced in a continuous melter provides additional flexibility and makes the waste forms easier to inspect. A small glass bead can also be heat-treated to produce semi-crystalline forms such as glass-ceramics.

Production of crystalline waste forms such as supercalcines and SYNROC normally involves drying chemically adjusted waste in a spray calciner as a first step. The dried waste may then be formed into beads using equipment such as the PNL disc-pelletizer shown in Figure 20. This equipment can produce crystalline waste form beads at rates exceeding 100 kg/h. Following pelletizing, the beads are sintered at 900⁰ to 1200⁰C to complete formation of the desired crystals.

In order to produce a waste product with improved stability, strength, and thermal conductivity, glass or crystalline beads may be encased in a metal matrix inside other highly inert barrier materials. Metal-matrix encapsulation equipment is now being tested at PNL on an engineering scale.

The entire system--from liquid HLW feeding through calcination, final-form production, encapsulation in metal matrix, and effluent treatment--is being combined in a total pilot-scale system. This flexible system allows production of several selected waste forms, during which equipment interactions and total system performance can be evaluated and improved.

FIGURE 20. Disc-Pelletizer

QUALITY VERIFICATION

Verifying the quality of large-scale canisters filled with solidified waste requires special techniques and process control. A program is under way that is intended to develop nondestructive tests and improved methods for confirming the quality of the waste form inside the canister. In this program, samples cored as shown in Figure 21 from full-scale solidified waste specimens are compared with laboratory glass samples to determine the effects of large-scale operations. This testing will provide data for plant operators and licensors regarding conditions that produce a high-quality waste form.

FIGURE 21. Core-Drilling of Full-Scale Canisters Filled with Simulated Waste

CANISTER DEVELOPMENT

To provide a containment barrier and to facilitate handling, solidified HLW is encased in a metal canister. Depending on the conditions to be encountered, the canister may be made of mild steel, stainless steel, or more exotic alloys. Each carefully designed canister that is filled with glass or an alternative waste form at PNL is fully analyzed and characterized. Canister designs are based on engineering-scale operating experience.

Once filled with solidified radioactive waste, the canister is sealed, leak-checked and decontaminated. Systems for performing these functions, such as the remotely operable welder shown in Figure 22, are being developed for use in an immobilization facility.

FIGURE 22. Remotely Operable Canister-Closure Welder

RADIOACTIVE PROCESS DEMONSTRATIONS

Large radiochemical engineering cells at PNL contain processing equipment for feeding, calcining, vitrifying, and treating effluents from solidification of actual HLW. This equipment is remotely operated and maintained, yet highly flexible. It has been used in two major demonstration programs: the Waste Solidification Engineering Prototypes Program and the Nuclear Waste Vitrification Project.

WASTE SOLIDIFICATION ENGINEERING PROTOTYPES PROGRAM

Two of the processes developed in the High-Level Waste Immobilization Program have previously been tested on a pilot scale with actual HLW during the Waste Solidification Engineering Prototypes Program. This extensive test and development program, conducted from 1966 to 1970 at PNL, demonstrated several waste solidification processes, including:

- pot calcination
- spray calcination
- continuous metallic-melter vitrification
- in-can melting.

Calcine, phosphate glass, phosphate-ceramic and borosilicate glass were produced. The spray calciner, continuous metallic melter, and in-can melter used in the program are shown in Figure 23.

In all, 33 engineering-scale runs were made using actual HLW at rates of 10 to 20 liters of liquid waste per hour. This waste, containing some 50 million curies of radiation, was solidified in thirty-three 8-ft-tall canisters with diameters of 6, 8 and 12 inches. Off gas from these runs was processed through a series of wet-scrub steps and filters, which reduced the radionuclides in the gas to levels well below the established federal standards.

This program covered a wide variety of processing conditions, utilized fully and semi-remote operating techniques, and formed the foundation of the recent program to demonstrate the vitrification of actual power reactor HLW.

NUCLEAR WASTE VITRIFICATION PROJECT

The Nuclear Waste Vitrification Project was conducted from 1978 to 1979. Spent power reactor fuel for this project was brought to PNL facilities; it was then sheared, dissolved, and the HLW was extracted to provide the feed for two spray calciner/in-can melter demonstrations. Two 8-in.-dia by 8-ft-tall canisters of waste glass were used. One of these canisters is shown in Figure 24. Each canister contains the waste from more than one metric ton of uranium fuel--about a million curies of radiation and four kilowatts of decay heat.

With the completion of the Nuclear Waste Vitrification Project in early 1979, the facilities were made available for further pilot-scale waste immobilization testing.

FIGURE 23. In-Cell Waste Immobilization Equipment

FIGURE 24. Canister of Actual High-Level Waste Glass

SMALL-SCALE IMMOBILIZATION TESTING

The spray calciner/in-can melter system shown in Figure 25 has been installed in shielded cells for testing with actual HLW. This system is situated in a cell adjacent to the pilot-scale equipment cell described previously. This scaled-down system operates at liquid rates of 0.2 to 2.0 ℓ/h . At this rate many reasonably long tests can be conducted with the available supplies of liquid HLW. Nonradioactive testing shows that this small-scale system accurately simulates many features of the plant-scale calciner/melter system. Effluents from this system are characterized and various treatment techniques are evaluated using actual defense and commercial HLW. Evaluation of equipment response to off-standard conditions is obtained rapidly and relatively inexpensively in this system.

The in-cell calciner will be used with selected alternative waste form processing equipment to test process flowsheets and to produce actual HLW samples under representative conditions. This equipment is designed in modules to facilitate replacement of the in-can melter with a pelletizer, a press, or other crystalline waste form processes. Analysis of the waste form, process effluents and operating parameters will allow direct comparisons to the glass waste form, thus facilitating appropriate waste form selections.

Figure 25. Small-Scale Immobilization Equipment

CONCLUSIONS

The High-Level Waste Immobilization Program is providing technology to allow safe, affordable immobilization and disposal of nuclear waste. Waste forms and processes are being developed on a schedule consistent with national needs for immobilization of high-level wastes stored at Savannah River, Hanford, Idaho National Engineering Laboratory, and West Valley, New York. This technology is directly applicable to high-level wastes from potential reprocessing of spent nuclear fuel. The program is removing one more obstacle previously seen as a potential restriction on the use and further development of nuclear power, and is thus meeting a critical technological need within the national objective of energy independence.

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