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HIGH-LEVEL RADIOACTIVE WASTE MANAGEMENT REVISITED

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INTRODUCTION

We prepared this paper as an overview of the technology available for managing high-level radioactive waste. We include a brief introduction to past and existing practices, and we list the alternative concepts under consideration for disposing of and isolating waste forms. We also discuss the U.S. vitrifitication program including waste formulation, process development and waste form characterization. In addition, we summarize the status of alternative waste form development. Finally, we offer our conclusions on the adequacy and acceptability of borosilicate glass as a first generation waste form, on the improved stability and protection that may be offered by alternative waste forms, and on the added protection that could be given waste in a repository through advanced engineering.

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BACKGROUND

Generation of high-level radioactive liquid waste began in late 1944 with the startup of chemical separation plants at the Hanford defense production facility. Since 1944, and even earlier during the research and development phases at Oak Ridge, Tennessee, the management of radioactive waste has been a prime concern. Procedures were developed to isolate the waste and to inhibit the release of radioactivity to the biosphere. Starting at Oak Ridge, and later continued in practice at Hanford and Savannah River, the radioactive waste generated as acidic solutions were neutralized and stored in underground tanks.[a] Improvements in reprocessing resulted in increased concentrations of radioactivity in the waste, which in turn required advances in tank design and waste management operations. Design and operation capitalized on the heat that is released from radioactive decay, by allowing the stored solutions to self-concentrate through evaporation to volumes as small as practicable.

It has long been recognized that storing high-level waste as aqueous solutions is only an interim solution to the problem of waste management, that the storage tanks have a limited life and could leak unless the contents are moved periodically to new and sounder tanks, and that the final treatment and storage of the waste should be as a solid. Since 1968 the approach at Hanford (shown schematically in Figure 1) has been to concentrate supernatants, in so far as feasible, to a salt cake, which is a less mobile medium than an aqueous solution. Prior to evaporation, cesium and strontium are removed from the solutions and sludges and are encapsulated for isolated storage. Beyond about five years after reactor discharge, the isotopes, ^{137}Cs and ^{90}Sr, are the major heat emitters. Noncrystallized, but saturated salt liquor is stored temporarily in new doubleshell tanks.[b]

(a) Mild steel was selected over stainless steel as tank material for reasons of availability and cost. Storage in mild steel tanks requires neutralization.

(b) These tanks are equipped with liquid detectors between the two tanks such that leaks in the primary tank can be detected and the contents transferred to a sound tank.

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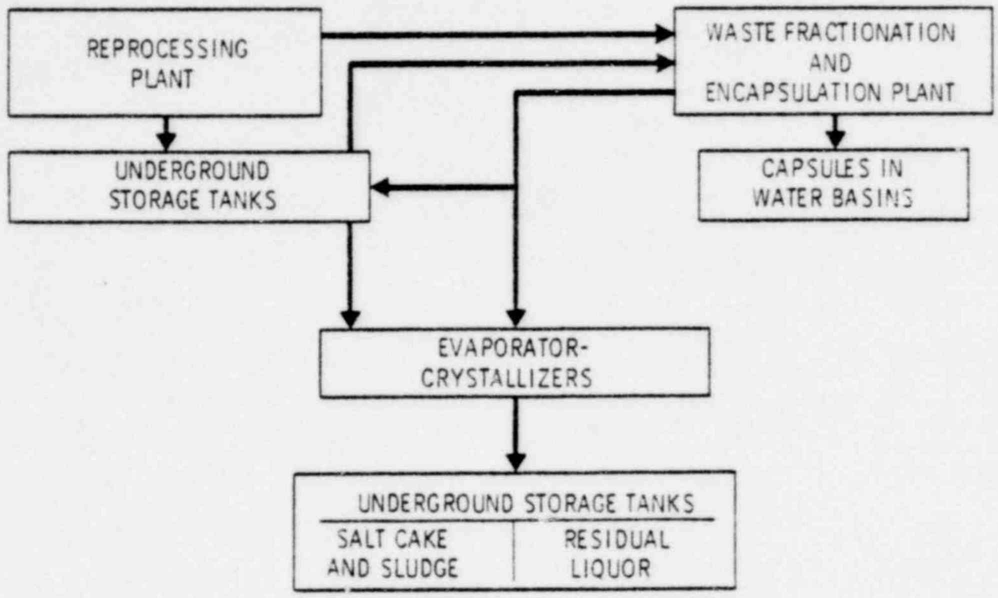

FIGURE 1. Simplified Schematic Flowsheet of Past/Current Hanford Waste Management

An extensive research and development program on processing and treating high-level liquid waste began in the U.S. in the early 1950s. The feasibility of converting acid waste to calcines and/or borosilicate or phosphate glasses was demonstrated at the Pacific Northwest Laboratory (PNL) in the late 1960s, using facilities provided as the Waste Solidification Engineering Prototype (WSEP). As will be noted later in this paper, these vitrification processes are well advanced and the latest commercial chemical reprocessing plant designs incorporate solidification technology for processing high-level waste. The Idaho Chemical Processing Plant (ICPP) at the Idaho National Engineering Laboratory (INEL) has for several years been converting its acid waste,[a] which is high in aluminum content, to a free-flowing calcine for storage in underground bins.

(a) From the beginning, because of the relatively low volume of its waste and because of the specialized nature of fuel processes at the plant, the ICPP stored acid waste in stainless steel tanks.

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The only existing commercial high-level liquid waste is that generated between 1966 and 1972 and still stored at the Western New York Nuclear Service Center, West Valley, New York. Management of most of that waste was by neutralization and storage in underground tanks, based on the technology available (from defense plant operations) at the time the West Valley plant was designed and constructed. A small quantity of acid waste containing unrecovered thorium from experimental commercial fuels is stored in a stainless steel tank in an underground vault at West Valley.

Existing waste management practice involves a continuing and extensive program of surveillance and monitoring of the waste. In addition, at the Hanford and Savannah River defense plants, the construction of new tanks of improved design and the transfer of waste from old to new tanks are required to reduce to a practicable minimum the probability of additional leaks and unplanned releases of radioactivity to the environment. To eliminate this unrelenting need, the processes shown in Figure 2 have been developed to

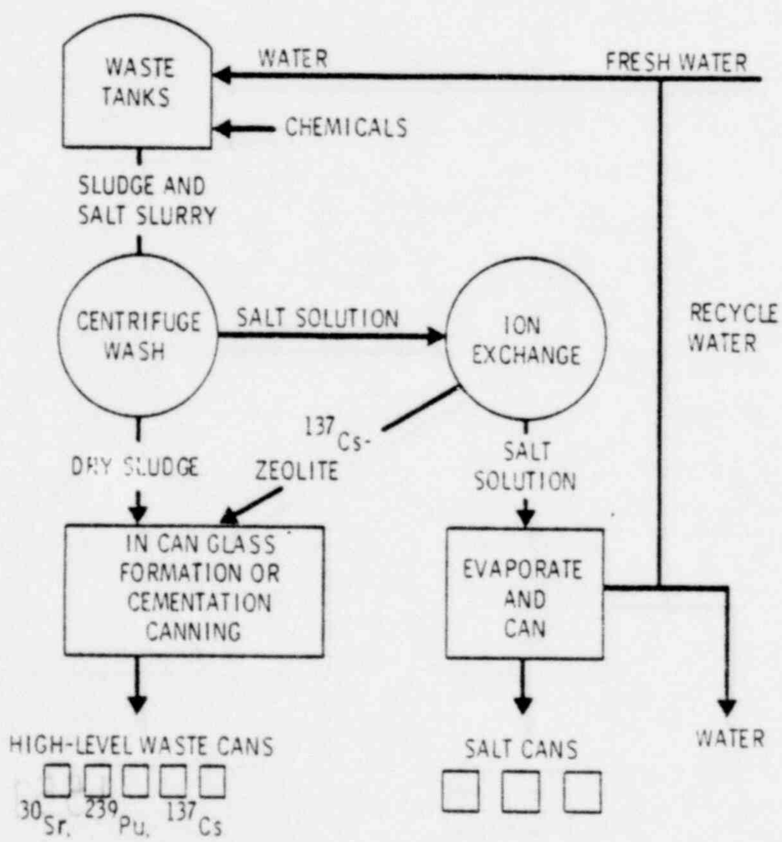

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FIGURE 2. Conceptual Waste Solidification Process

isolate and process the radionuclides in the waste into stable solids that are suitable for disposal in Federal repositories isolated from the ecosystem. Borosilicate glasses have been proposed for the first generation of high-level waste solids.

Of significance to this symposium is the observation that no measurable impact on the health and safety of human populations has resulted, either from past waste management practices, which have resulted in unplanned releases (leaks) from underground storage tanks, or from other waste management activities, including the intentional storage of radionuclides in Hanford soils.

ADVANCES IN HIGH-LEVEL RADIOACTIVE WASTE MANAGEMENT

The objective of an advanced high-level radioactive waste management program is to convert the waste to a form that may be placed in a location that permanently isolates the waste from the human biosphere. A partial listing of options that have been and are under active consideration for disposing of the waste is given in Table 1. Ten options were presented in the recent environmental impact statement on management of commercially generated radioactive waste. A discussion of the pros and cons of these options is outside the scope of this paper; remarks relating to the permanent disposal of radioactive waste will be limited to the current leader, i.e., deep geologic disposal, since deep geologic disposal could be chosen for pilot plant demonstrations based on existing and near term technology.

Over the past years, many different waste forms have been proposed and/or developed to varying degress for the immobilization of high-level nuclear wastes. These waste forms range from fairly simple materials, like concrete and glass, to more advanced forms such as sintered ceramics, glass-ceramics, hot-pressed ceramics, and matrix materials. The number of proposed waste forms has greatly increased in recent years and there is a general trend toward the more advanced forms having multiple layers or barriers for protection. The advanced forms are more complex, less developed, and more

TABLE 1. Options for Disposal of High-Level Radioactive Waste

- Deep Geology
  - Salt Beds
  - Salt Domes
  - Granite
  - Basalt
  - Tuffs
  - Shale
- Seabed
- Antarctic
- Space

expensive to produce. Because the environment (temperature, pressure and composition of solids or fluids that the waste may contact) to which the waste forms will be exposed will vary greatly with the storage system, it is very important that the entire system be specified before a waste form is selected. However, we believe that the properties of some waste forms are sufficiently understood such that once the storage conditions are fixed, a waste form can be specified that will satisfactorily contain the radioactivity.

METHODS FOR DISPOSAL AND ISOLATION

What is the purpose of geologic disposal? Figure 3 schematically depicts the multiple barrier isolation concept. This concept utilizes a series of barriers, each designed to perform a specific function. Use of multiple barriers is standard in the nuclear industry and has contributed to its outstanding safety record. For a geologic repository the multiple barriers will consist of: 1) remoteness from the biosphere, 2) specially selected near-field geologic properties, such as dryness and impermeability, 3) specially designed engineered barriers, including backfill materials with sorptive

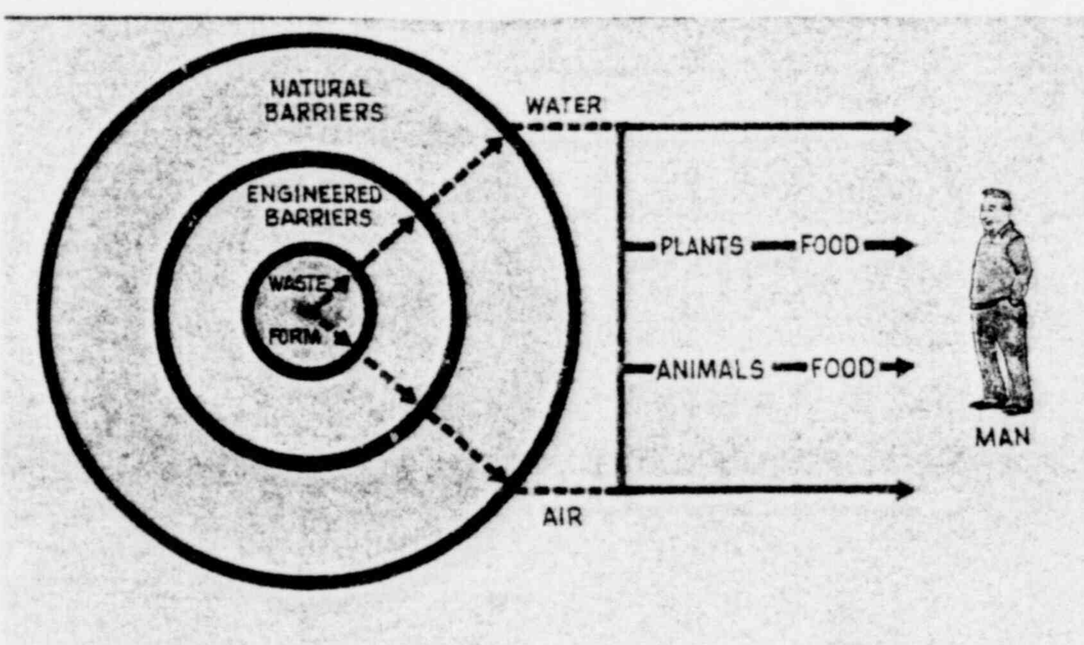

FIGURE 3. Multiple Barrier Isolation Concept

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properties (optional), 4) an overpack (optional), 5) the canister, and 6) the solidified waste form. It is important to note here that the waste form is only one of a minimum of four barriers in the system. The optional use of overpacks and additional engineered barriers can easily increase the total number of barriers in the repository system to six or more.

Another important consideration is the transport of the waste from the site of its formation to the site of disposal. For this step the multiple barriers will be: 1) the waste form, 2) the canister, 3) an overpack (optional), and 4) the transfer cask. The design and licensing of waste casks should be an easy extension of the licensing of fuel shipping casks.

Figure 4 puts the task of disposing of high-level waste in perspective. It shows that after about 2000 yr the toxicity due to the radioactive materials in a repository of commercial high-level waste will begin to resemble the toxicity of a similar volume of average uranium ore. The figure also shows

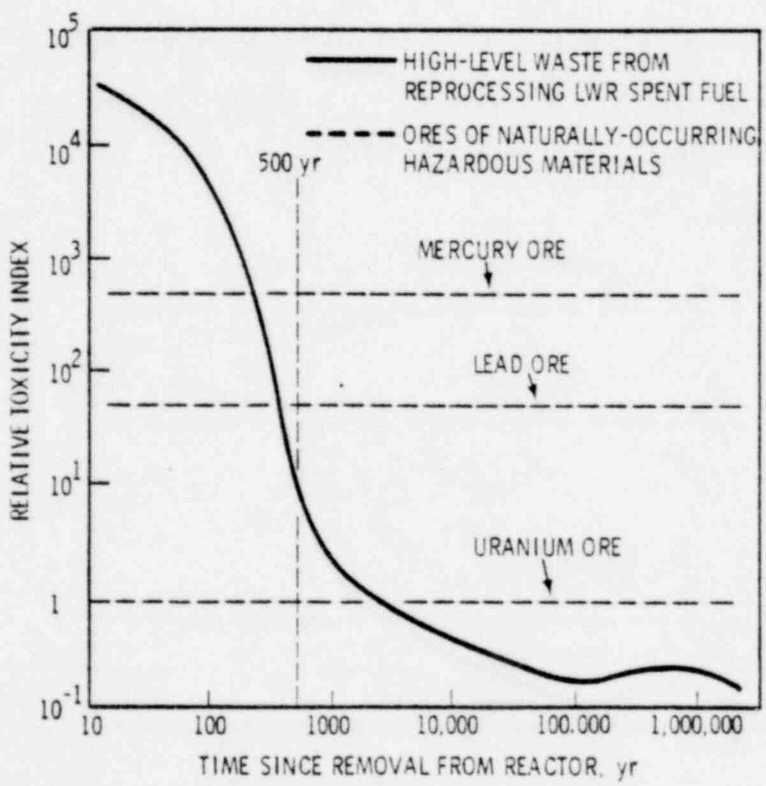

FIGURE 4. Relative Toxicity of Nuclear Waste

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that after 500 yr the hazard potential associated with the radioactive material in the repository is actually lower than the chemical hazard potential associated with some naturally occurring nonradioactive ores. Solidified defense waste, with its lower specific activity, would present an even lower toxicity hazard.

Repository temperature is a design variable. The heat generated in the repository can be controlled by a number of mechanisms. Less waste can be included in a canister either by diluting the waste with nonradioactive additives or frits, or by using a smaller diameter canister; fewer canisters can be deposited per unit area of repository; or, the waste can be stored for a period of time in a surface facility to allow major decay of the radioactivity. The temperature of the wall of a canister of high-level waste is plotted in Figure 5 as a function of time under two sets of conditions. The upper curve shows the temperature based on a "maximum efficiency design" for commercial high-level waste, i.e., 40 canisters per acre of repository area

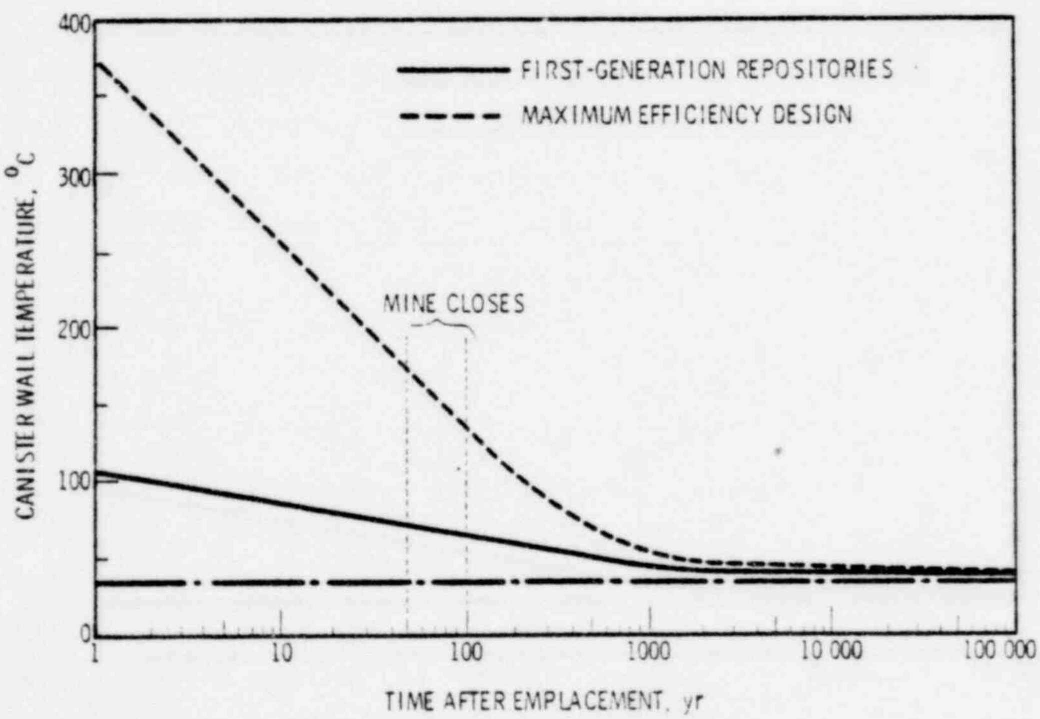

FIGURE 5. High-Level Waste Canister Wall Temperatures in a Salt Repository

with each canister generating about 3.5 kW of heat. The lower curve is representative of temperatures expected from solidified defense waste. The lower curve could also represent one or a combination of the more conservative approaches for commercial waste.

The temperatures shown in Figure 5 are for a dry repository, which has the properties, such as dryness and impermeability, that we would look for in our search for a repository. If water should somehow intrude into the dry repository, the heat transfer at the canister wall would increase and the wall temperature of the canister would drop significantly...to almost one-half the temperatures for the maximum efficiency design shown in the figure. Thus, the combination of high temperature in the presence of water is not expected, and in any event can be designed around.

An additional natural layer of defense, in the event water enters the repository and attacks the waste, is the leachate/geology interactions. The choice of the repository locations will include the expectation of sorption by and/or chemical reaction with the surrounding host rock, thus impeding (retarding) the movement of dissolved radionuclides with the water flow.

Engineered barriers for the waste package could include overpacks of highly resistant and stable metals (titanium, copper, lead) or ceramics (alumina, titania). Additional barriers can be added to the repository as backfills (clays, silica) to restrain movement of water to the waste, and to sorb radionuclides if released from the waste.

VITRIFICATION OF RADIOACTIVE WASTE

The waste form most fully developed is produced by calcining high-level liquid waste, mixing the calcine with a glass frit, and melting the mixture to form a monolithic glass. Although this can be accomplished by several processes, the most fully developed process involves continuous spray calcination and in-can melting. In this process high-level liquid waste is calcined in a hot tower, glass frit is added to the calcine, and the mixture is melted in a canister as shown schematically in Figure 6. This process has been tested at

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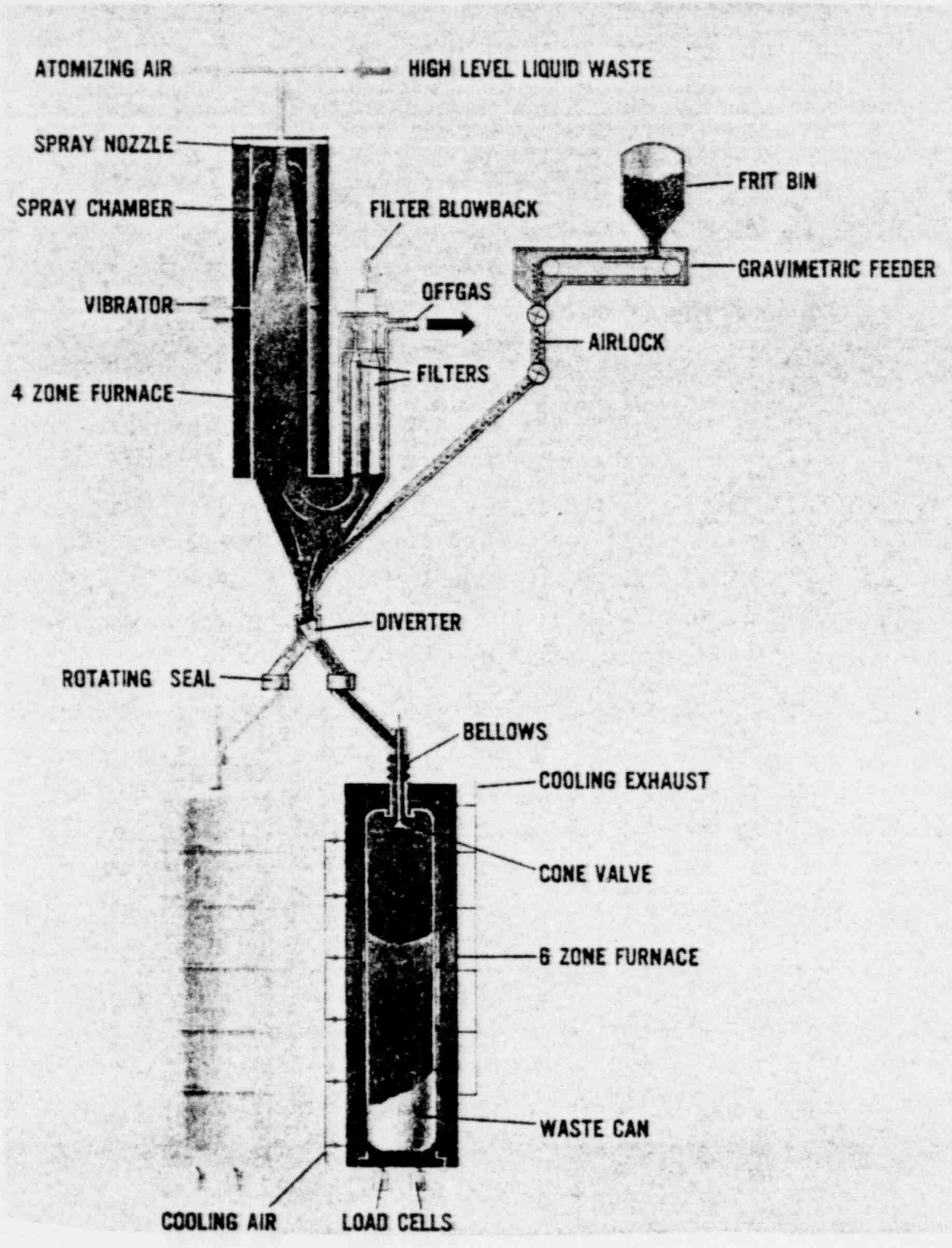

FIGURE 6. Schematic of Spray Calcination/In-Can Melting Process

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full scale on simulated, nonradioactive waste solutions, and the glass product has been evaluated. Figure 7 is a photograph of a full-scale nonradioactive pilot plant facility using this spray calcination/in-can melting process. Because the high-level liquid waste may have a range of compositions depending on the source (Savannah River, Hanford, Idaho, West Valley in New York, and proposed commercial reprocessing plants) many compositions have been tested. The spray calcination/in-can melting process has been shown to be adaptable to all of these waste compositions by adjusting the frit, additives, and processing conditions. PNL has demonstrated the process at near full scale using actual radioactive waste produced from commercial light water reactor fuel. Figure 8 is a picture of one of the canisters from the full-level radioactive runs completed under the liquid waste vitrification project earlier this year. A variation of the spray calcination/in-can melting process has been operated by the French at their Marcoule plant. A rotary kiln is used to form the calcine, frit is added to the calcine, and the mixture melted and cast into containers.

A second process, in which a continuous ceramic melter is used to vitrify waste, has not been as fully developed as the spray calcination/in-can melting process. The continuous melting process may be able to produce higher temperature glasses. In the continuous melting process, calcine from a spray calciner is fed along with the frit to the ceramic melter as shown in Figure 9. The ceramic melter is heated by passing an electric current through the glass. A photograph of the ceramic melter is shown in Figure 10. The continuous melting process has been tested at full scale using simulated, nonradioactive feed. The unit is capable of handling feed from a commercial reprocessing plant at a rate equivalent to that required for a five-ton-per-day capacity. A modification of the continuous melting process in which liquid high-level waste is fed directly to the ceramic melter, thereby eliminating a separate calcining step, has also been tested at full scale.

The spray calcination/continuous melting process is the preferred process system selected by the Savannah River Laboratory (SRL) for defense waste. An engineering-scale unit of this system has been operated at PNL using SRL

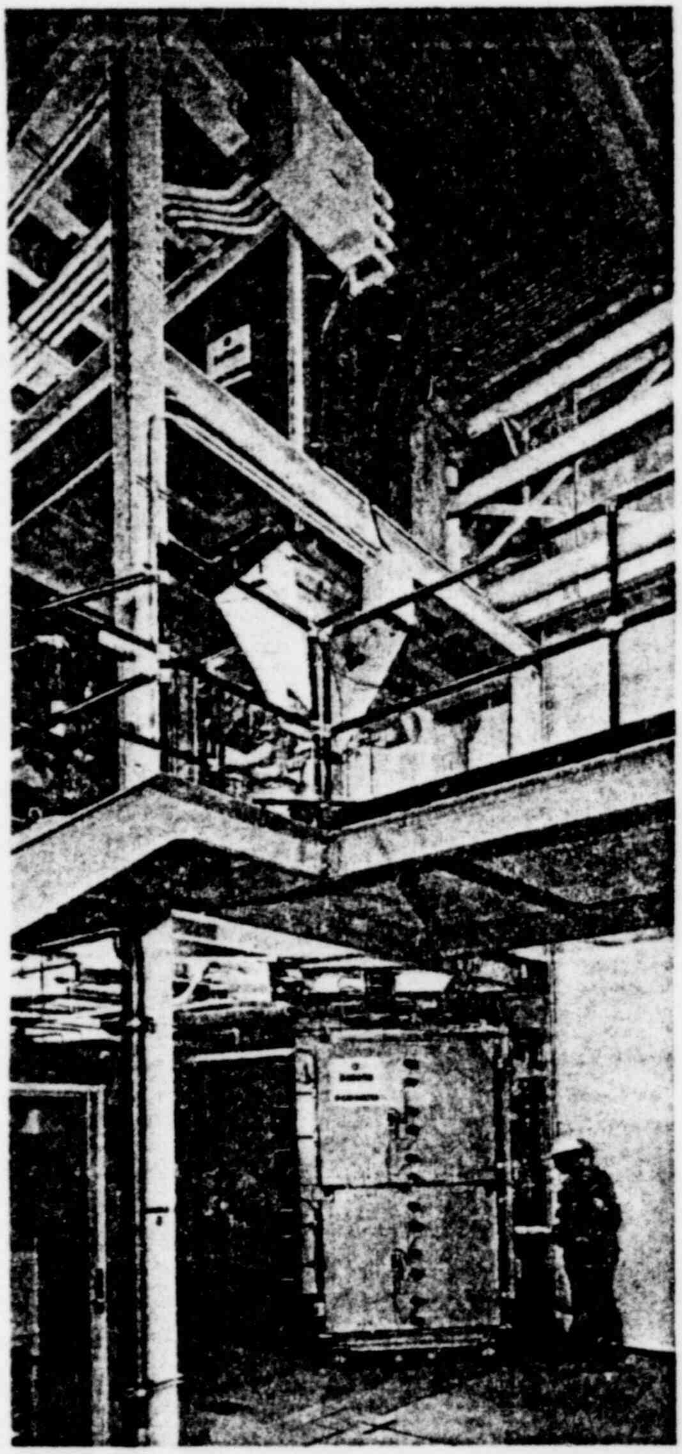

FIGURE 7. Spray Calciner/In-Can Melter Installation in PNL Nonradioactive Pilot Plant

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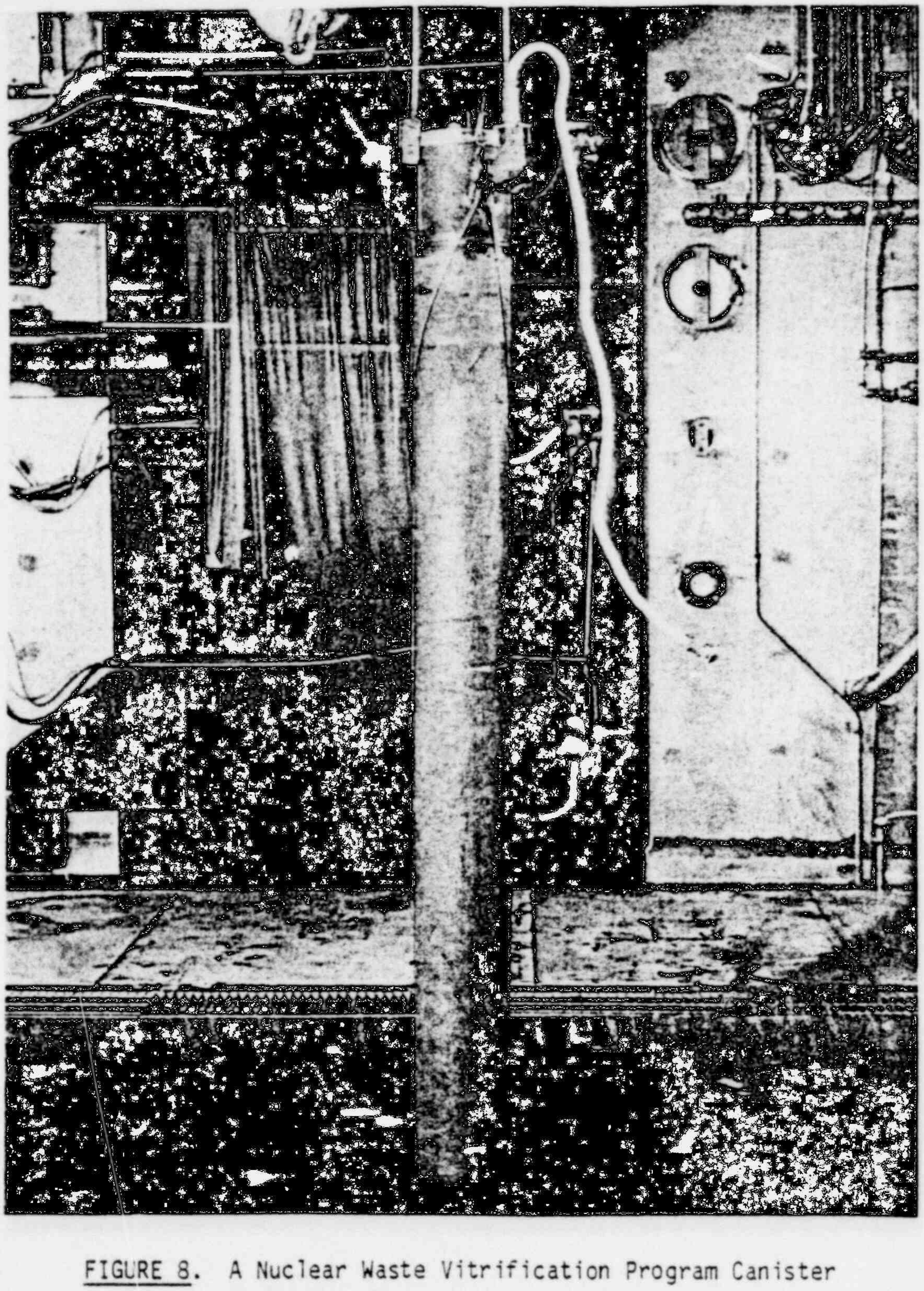

FIGURE 8. A Nuclear Waste Vitrification Program Canister

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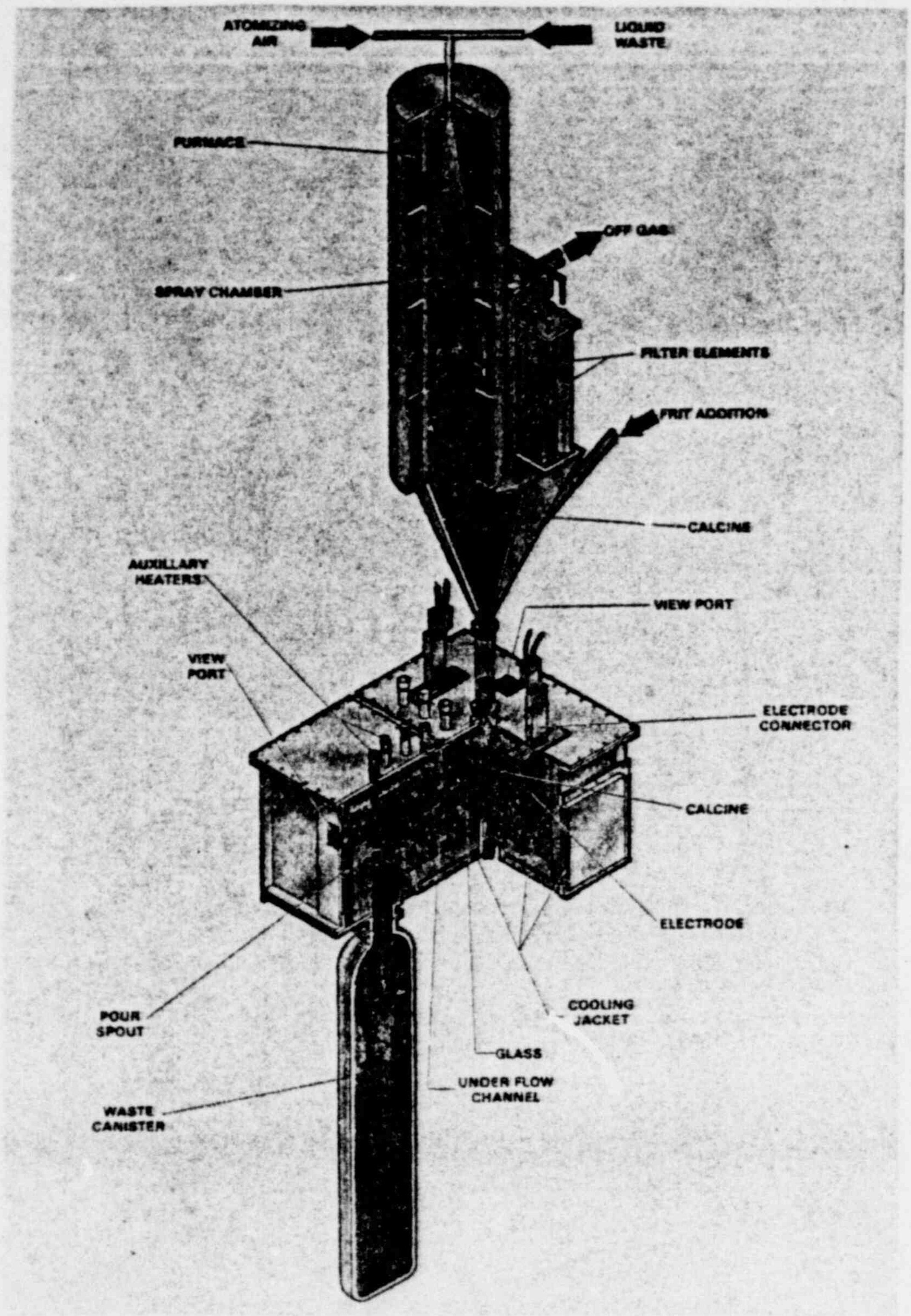

FIGURE 9. Schematic of Spray Calciner/Ceramic Melter System

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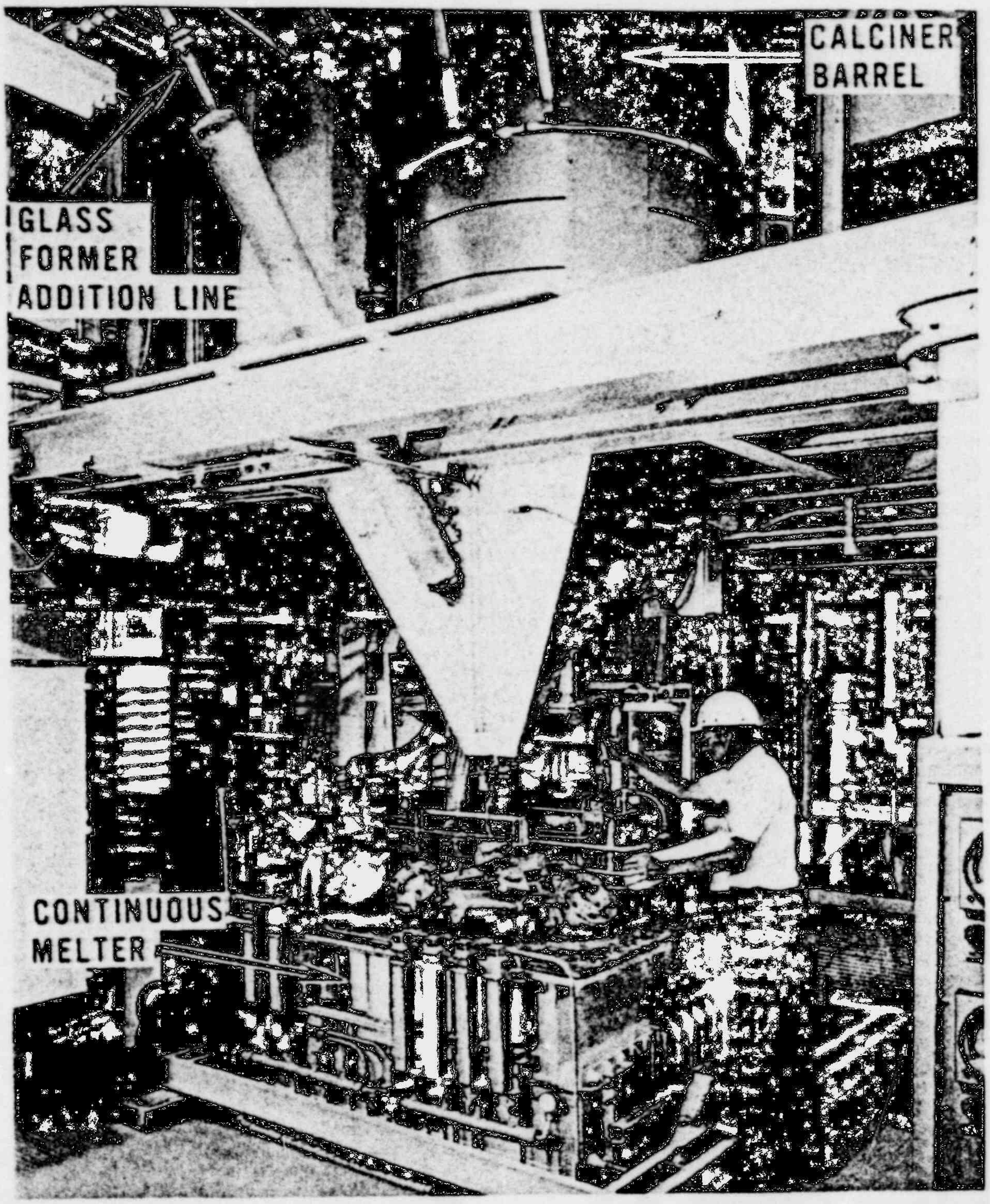

FIGURE 10. Spray Calciner Coupled to the Continuous Melter

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simulated waste. The feed rate to the spray calciner was 425 L/h and the continuous melter produced glass at a rate of 85 kg/h. Long-term runs, totaling about 40 days, using simulated, nonradioactive feed solutions have been completed at PNL to demonstrate the reliability of the spray calcination/continuous melting equipment. If it is decided that the continuous melting process is to be used to solidify the Savannah River or West Valley, New York wastes, the next step will be to design and test remotely operable equipment.

PROPERTIES OF VITRIFIED WASTE

The properties of the waste glasses produced by various solidification processes must be known in order to predict the behavior of the waste glasses during shipping, storage and disposal. For example, in the event that water should penetrate to the repository, a most important property is the rate at which the glasses dissolve under potential storage conditions. Both the overall leach rate (grams glass dissolved per cm^2-day) of waste glasses and the depletion of various isotopes have been studied as a function of different leaching conditions. Devitrification, which occurs when glasses are heated in the range of 500^o to 900^oC, increases the overall leach rate, but usually by no more than a factor of 2 to 5 (see Figure 11). The increased leaching may occur in the residual glass phase due to the depletion of silica, or it may occur in one of the crystalline phases that is formed. Other factors affecting leach rates are given in Table 2. Leach rates are measured either by determinating weight loss or by analyzing for leached ion concentrations in the leachate. At 25^oC measured leach rates are generally in the range of 10^{-4} or 10^{-7} grams of glass per square centimeters per day. Alkali and alkaline earth ions generally leach at rates one or two orders of magnitude higher than cations of higher-valence elements. It has been calculated that less than 1% of the radioactivity in glass would dissolve each thousand years, even if the glass were exposed to flowing water at ambient repository temperatures of 40^oC. Experiments have shown that even less radioactivity would dissolve in slow-flowing or stagnant water.

The primary leaching mechanism below about 80^oC is a diffusion-controlled ion exchange process in which hydrogen or hydronium ions exchange

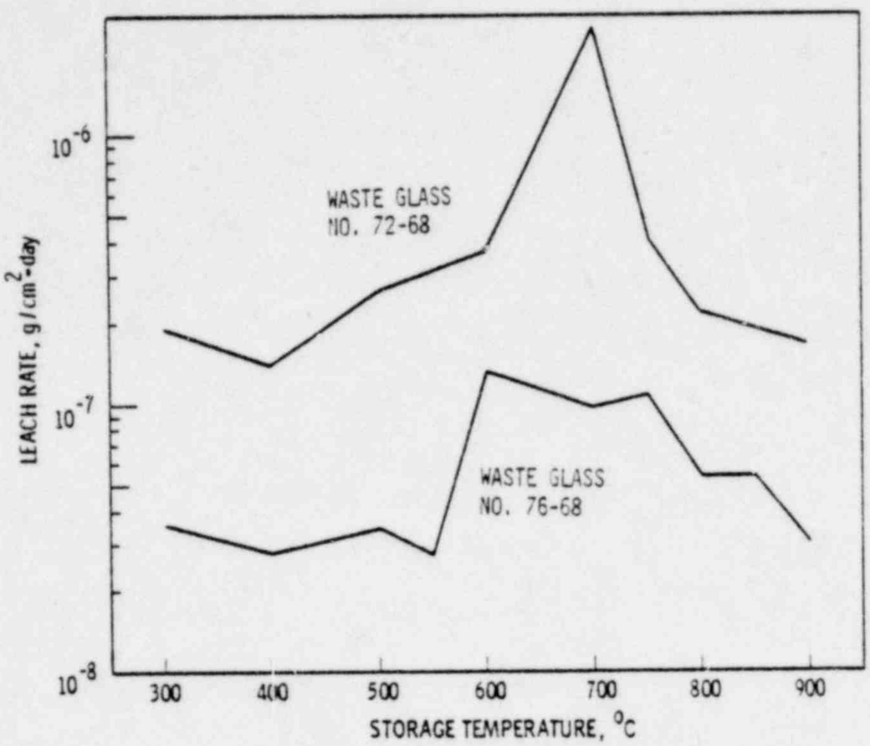

FIGURE 11. Leach Behavior as a Function of Devitrification for Two Representative Borosilicate Waste Glasses

TABLE 2. Factors Affecting Leach Rate Measurements

Intrinsic	Extrinsic
Glass Composition	Temperature
Thermal History	Pressure
Radiation Effects	Composition of Leachant
Physical Form	Flow Rate of Leachant
Surface Character	

for cations in the glass lattice. The resulting surface area of hydrous silica, depleted of metallic cations, provides a protective barrier that retards further leaching. At higher temperatures surface corrosion predominates. The leach rate increases roughly by a factor of 10 to 100 with each 100°C rise in temperature. However, as discussed earlier, there are

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several methods available for assuring that the temperature of the waste glass remains low so that the leach rates are acceptable.

Many other properties of waste glasses have been studied including viscosity, softening point, density, friability, thermal conductivity, thermal expansion, volatility, and transmutation. In addition, the long-term effects of self-contained alpha radiation have been simulated by doping the glass with ^{244}Cm and allowing the glass to receive a self-radiation dose equivalent to 500,000 yr of storage. The principal effect observed was the buildup of less than 60 cal/g of stored energy and a change in density of less than 1%. These effects are not considered to be a problem. As shown in Figure 12, the self-radiated glass did not fracture or change in appearance.

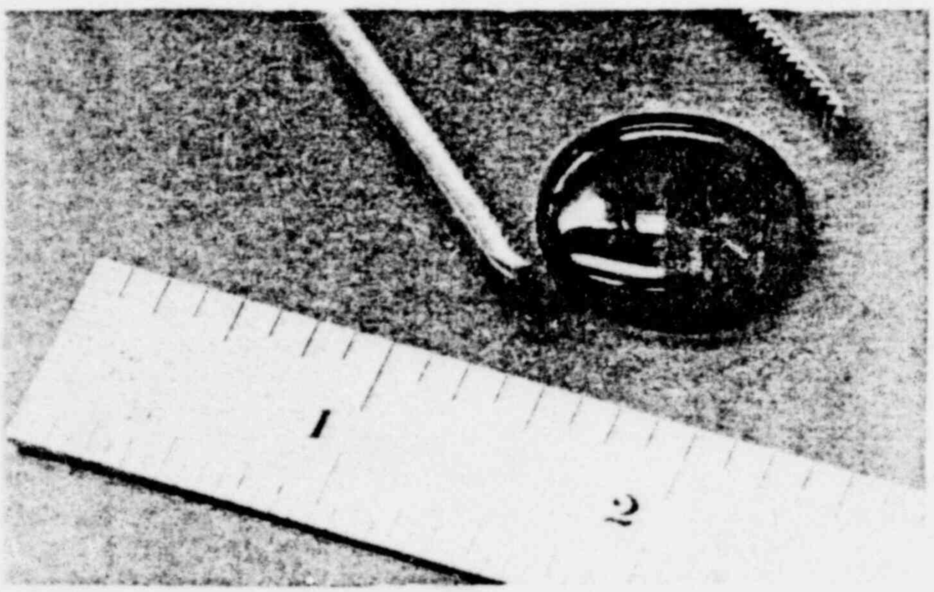

FIGURE 12. Waste Glass Self-Radiated to Equivalent of 500,000 yr of Storage

ALTERNATIVE WASTE FORMS FOR RADIOACTIVE WASTE

Although it is the general consensus of the worldwide technical community that glass is an acceptable waste form when used in a geologic storage system, other waste forms are being investigated. Each waste form, whether it is

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glass, ceramic, crystalline, or matrix has some advantages when a single property is considered. Although the processes for the alternative waste forms are generally more complex to engineer, especially for remote operation, and are likely to be more expensive to produce, they may be more resistant to leaching, have reduced volatility of some radionuclides and have greater impact resistance. Some of these alternative waste forms are listed in Table 3. The broad categories of these waste forms are discussed briefly below.

Concretes and Grout

Concretes and grout have a long history of practical application. Cement mortar produced by the Romans has held stone aquaducts together so well that they can still be used after 2000 yr. Concrete and grout technology has also

TABLE 3. Alternative High-Level Radioactive Waste Forms

Concretes and Grouts	Hot-Pressed Ceramics
Cement and liquid waste	Hot-pressed calcine
Cement and calcine	Hot-pressed supercalcine
Cement and supercalcine	Hot-isostatic-pressed calcine
Cement and sludge	Hot-isostatic-pressed supercalcine
	Hot-pressed concrete
	SYNROC B
Sintered Ceramics	**Coated Particles**
Supercalcine	Glass cores: Glass coating
Sintered calcine	Crystallic cores: PyC/SiC coatings
Sintered titanate	Crystalline cores: PyC/Al_2O_3 coatings
Clay-based products	Crystalline cores: PyC/SiO_2 coatings
SYNROC A	
Glass-Ceramics	**Metal Matrices**
Celsian glass-ceramics	Glass marbles / Metal matrix
Recrystallized fusion melts	Sintered-ceramic cores / Metal matrix cermets
Basalt glass-ceramics	Coated glass marbles / Metal matrix
	Coated supercalcine cores / Metal matrix

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been applied to low-level radioactive immobilization. Commercial firms supply equipment and processses for concreting reactor radioactive wastes. Concentrated low-level wastes are routinely mixed with grout and pumped into an underground shale formation in the hydrofacture waste disposal process used at the Oak Ridge National Laboratory (ORNL). Concrete immobilization of the Savannah River Plant wastes has been investigated, and it has been considered for commercial high-level wastes. The ORNL is investigating the use of concrete formed under elevated temperature and pressure (FUETAP). However, in addition to the need for developing a remotely operable process and defining optimum concrete formulations for various waste compositions, there are unresolved questions concerning the thermal and radiation stability of concrete.

Sintered Ceramics

Sintered ceramics are prepared by mixing calcine with glass frits, clays and other minerals, and consolidating them by subsequent heat treatment. The resulting product is principally crystalline, although some amorphous phases may be present. Consolidating sintered ceramics may involve pressing or pelletization prior to heat treatment. The best known of these waste types are the supercalcines, which have been developed by Dr. Gregory J. McCarthy and co-workers at Pennsylvania State University under a subcontract administered by PNL under a DOE program. Other variants are synthetic minerals that are produced by crystallizing ceramics from melts of the waste calcine mixed with suitable raw materials, as proposed by Dr. A. E. Ringwood of Australia.

Glass-Ceramics

Glass-ceramics are formed by the controlled crystallization of glass melts, using a heat treatment process involving nucleation and crystal growth. Dr. W. Lutze and associates at the Hahn-Meitner Institute in West Germany have been the leading investigators in the incorporation of radioactive waste materials in glass-ceramic waste forms, and have conducted extensive studies on the properties of this waste form.

Hot-Pressed Ceramics

Hot-pressed ceramics may be formed by a number of approaches, with or without additives. The FUETAP concretes may be placed in this classification.

The second generation of synthetic minerals proposed by Dr. Ringwood (SYNROC B) are of this class. Supercalcines may also fit this group.

Engineering development of processes to produce the hot-pressed ceramics, as well as processes to produce sintered or glass-ceramics, is just getting underway. Generally, the ceramic forms are more difficult to formulate and produce than glass. However, the crystalline, ceramic forms are thermodynamically more stable than glasses, although some crystalline materials are known to undergo metamictization, i.e., conversion to amorphous materials in the presence of radiation. Some forms, particularily the SYNROC B synthetic minerals, are claimed to be much more resistant to leaching by water at high temperature under high pressure.

Coated Particles

Coated particles can provide an additional barrier for the radionuclides in a waste form. Coatings are selected for their inertness and chemical durability. Pyrolytic carbon coatings are used for crystalline waste forms, with an overcoat of alumina, silica or silica carbide. Multibarrier waste forms can be prepared in the laboratory. The remote engineering processes for coating would be complex.

Metal Matrices

Coated or uncoated beads, marbles or particles could be encapsulated in a metal matrix. A multibarrier waste form of coated supercalcine in a metal matrix is being developed at PNL. A cermet waste form has also been developed at ORNL. It consists of a continuous iron-nickel base metal containing small particles of radioactive waste oxides.

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CONCLUSIONS

A geologic repository is currently the leading option for disposing of radioactive wastes. Among the design objectives to be considered in the selection of a site for a geologic repository are the dryness and impermeability of the geologic formations. In addition, should water unexpectedly intrude into the repository and leach radioactivity from the waste, the surrounding host rock should have geochemical properties that permit favorable interaction with the leachate.

After a period of 500 yr the hazard potential of the waste form reduces to a level comparable to that of natural ores containing toxic metals, and after about 2000 yr the hazard potential becomes equivalent to that of uranium ores.

Several vitrification processes have been developed to produce borosilicate glass waste forms. The borosilicate glasses have been shown to have acceptable leach rates in the event that they are exposed to water under anticipated storage conditions. They have also been shown to be physically stable after having received an alpha radiation dose equivalent to 500,000 yr of exposure. The canister containing the borosilicate glass adds a barrier impeding the access of water to the waste form; and if needed, additional barriers can be provided by overpacks and the addition of selected backfill materials to the geologic formation.

Among potentially improved waste forms, there are coated particles, superior calcines, synthetic minerals and cermets. Generally the ceramic forms are more difficult to formulate and produce. However, because there may be better waste forms than glass when some individual properties are considered, additional studies of the more complex forms are being performed. Engineering development of the processes for these forms is just getting underway.

Borosilicate glass is generally accepted by the technical community as a suitable waste form that could be selected now for disposal in a deep geologic repository. On the otherhand, it should be noted that no single known waste form offers optimum performance over the entire range of properties to be considered. Either glass or crystalline materials have some advantages when

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only a single property is considered. However, while there is no best waste form, as there may not be a best geology, there certainly are several, if not many, acceptable combinations of waste forms, added barriers, and geologies.

Sound technology is available to protect the health and safety of the public from exposure to the radioactive materials, not only during the processing of high-level nuclear wastes, but also during the shipment of the packaged waste, during the emplacement of the waste in a repository, and finally for thousands of years and beyond. The alternatives for combinations of waste forms and geologies are many (perhaps too many). The decision makers may select one of several suitable and adequate alternative waste disposal methods. Input from the public will contribute to the value judgments required to determine the degree of conservatism needed to be incorporated in a waste management system; but the technology for making a decision is available now, and a decision based on technology can be made now.

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