DOE/EA-0179

Environmental Assessment

Waste Form Selection For SRP High-Level Waste

July 1982

U.S. Department of Energy
Assistant Secretary for Defense Programs

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1. SUMMARY

The Department of Energy (DOE) has recently decided to construct and operate a Defense Waste Processing Facility (DWPF) at the Savannah River Plant (SRP) to immobilize the high-level radioactive waste generated and stored pending disposal in a federal geologic repository. The Savannah River Plant is a major installation of DOE for producing nuclear materials for national defense. About 110,000 m^3 (28 million gallons) of high-level waste are now in storage in underground tanks at SRP*. The immobilized waste from the DWPF will be the initial barrier of the proposed multibarrier engineered and geologic system for disposal of high-level waste. Based on the recently completed waste form screening program, DOE has the necessary data to select the waste form for the DWPF. The purpose of this document is to assess the potential environmental consequences of selecting borosilicate glass as the immobilization form for high-level waste at SRP.

In the immobilization process the high-activity fraction of the SRP high-level waste is mixed with glass frit to form the feed for the melter. The glass is cast from an electric-heated, ceramic-lined melter into canisters 0.61 m in diameter and 3.0 m high. The molten glass solidifies into a chemically inert, highly insoluble, nondispersible, nonvolatile solid with very low measured leachabilities in simulated groundwater. Thermal stability and structural stability against self-irradiation effects of the glass form are fully sufficient to maintain waste form integrity. Key properties of the borosilicate glass waste form are shown in Table 1-1.

The borosilicate glass form, within the proposed multibarrier waste disposal system, contributes to the isolation of the waste from the accessible human environment. Borosilicate glass has sufficient mechanical strength and impact resistance to resist the stresses of repository emplacement (and retrieval during a specified retrieval period). It is compatible with a full range of repository geologies, and has projected (fractional) release rates into repository groundwaters of less than 1 part in 10,000 per year, as required by proposed DOE waste form specifications.

* The waste is composed of insoluble sludge, precipitated salts, and supernatant (liquid). The actual volumes at any time in the future will be a function of the waste generation from plant operations, DWPF startup, and the operations of processes to concentrate the waste.

TABLE 1-1

Key Properties and Characteristics of Borosilicate Glass Waste Form

Property or Characteristic	Borosilicate Glass
Density, g/cm^3	2.75
Waste Loading, wt %	28
Toleration of Waste Variability	Acceptable
Long-Term Leachability,* $g/m^2 \cdot d$	10^{-3} to 10^{-4}
Fractional Release Rate from Full-Size Form,** yr^{-1}	10^{-5} to 10^{-6}
Radiation Stability	Very good
Impact Response,† wt % fines	0.14 to 0.18
Processability††	Relatively simple

* Based on plutonium leach rates in long-term tests at room temperature.

** Estimated from plutonium leaching data (conservatively assumes that release of radionuclides is not reduced by solubility limitations).

† Generation of particles less than 10 micrometers in size from single impact of 10 J/cm^3 energy density.

†† Relative ease of producing the waste form.

Calculated doses and health effects from emplaced waste in potential repositories during the isolation period are small and are not significantly influenced by any reductions in leachability below current values for the borosilicate glass. Under most circumstances, peak doses are calculated to be less than 1% of the dose from natural background. For a typical repository, credible events which might damage the repository and its emplaced waste would not significantly affect this dose. The low solubilities of many of the radionuclides and their sorption on engineered barriers and on the surrounding rock should significantly reduce the release rates below those predicted from the leach tests and used in the repository consequence analysis.

Crystalline ceramic,* the leading alternative to borosilicate glass, also appears to be an acceptable form for immobilizing the SRP high-level waste. Both are expected to meet regulations and repository acceptance criteria. The assessment also shows that the environmental effects of disposing of SRP high-level waste as a crystalline ceramic form would not differ significantly from the projected effects for disposal of the borosilicate glass form. A comprehensive evaluation program led to the recommendation of borosilicate glass as the preferred waste form because process complexity, development requirements, and programmatic costs were determined to be less for borosilicate glass than for crystalline ceramic. The utilization of the borosilicate glass is supported by waste form evaluation programs in other countries in which essentially all other nations now reprocessing or planning to reprocess spent nuclear fuels are either using borosilicate glass or have selected borosilicate glass as the preferred high-level waste form.

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* Crystalline ceramic is a generic term for a product of compatible mineral phases, formed at high temperatures. Two candidate waste forms, Synroc-D (a titanate-based ceramic developed by Lawrence Livermore National Laboratory) and tailored ceramic (an alumina/rare earth-based ceramic developed by Rockwell International), are included in this term.

2. PURPOSE AND NEED FOR ACTION

2.1 PURPOSE

The purpose of this document is to assess the potential environmental consequences of the proposed action to select borosilicate glass as the waste form for the Defense Waste Processing Facility (DWPF). The DWPF will immobilize the high-level radioactive waste generated and stored at the Savannah River Plant (SRP).

Potential environmental impacts of an alternative waste form and its selection process are also included in this document.

2.2 NEED FOR PROPOSED ACTION

Since 1953, SRP has been producing special nuclear materials for defense purposes. Chemical separations of irradiated fuel and targets at SRP result in product streams and acidic liquid streams that contain almost all of the fission products and small amounts of transuranics. Currently, this waste is chemically converted to an alkaline solution and stored in large underground tanks at SRP as insoluble sludges, precipitated salts, and supernatant (liquid).

The Department of Energy (DOE) has initiated activities to dispose of the defense high-level waste generated at SRP. As part of the system approach, the high-level waste will be immobilized into a highly dispersion-resistant waste form;* canisters of the immobilized waste are to be later emplaced within multibarrier systems in deep geologic repositories. Construction of the DWPF, which will produce the waste form, is currently scheduled to begin in 1984; operation of the DWPF is scheduled to begin in 1990.

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* The DWPF will be constructed in stages.[1] First, the insoluble sludge (containing most of the strontium-90 and the actinides, and presenting the greatest long-term radiological hazard) will be immobilized. Next, radioactivity in the precipitated salt andm supernatant (primarily cesium-137, plus small amounts of strontiu and actinides) will be removed and either recovered for beneficial use or mixed with the sludge feed prior to immobilization. The current plan is to dispose of the decontaminated salt on the SRP site as low-level radioactive waste.

When conceptual design of the DWPF was started in 1977, borosilicate glass was selected as the reference waste form on the basis of extensive DOE studies. In FY-1979, the National High-Level Waste Technology Program was established to develop the technology for immobilizing high-level waste into solid waste forms which would provide highly efficient barriers against radionuclide release to the environment. Since the inception of the program, seventeen candidate waste forms have been developed and characterized by some fourteen participating contractors. Based on screening evaluations,[2,3] as well as on work at SRP and other laboratories, the number of forms under consideration was reduced from seventeen to seven. Further assessments culminated with the selection in November 1981 of two forms--borosilicate glass and a crystalline ceramic--for consideration as the final DWPF waste form.[4]

Based on data on waste form characteristics and expected repository performance, DOE is ready to select the final waste form for the DWPF. It is desirable to make the final waste form selection as early as possible to allow firm design of the DWPF, to reduce the scope of alternative waste form studies, and to increase efficiency by concentrating research and development on a single form.

2.3 RELATION TO OTHER PROGRAMS

2.3.1 Other U.S. Waste Form Programs

In preliminary evaluations[5,6] of waste forms for immobilizing Hanford and Idaho high-level waste, borosilicate glass and crystalline ceramic forms were consistently ranked among the top candidate waste forms. Borosilicate glass is the reference form for the high-level waste at West Valley, but other alternatives are being examined.[7] The program to select a waste form for future commercial high-level waste is being developed.

2.3.2 Other High-Level Waste Disposal System Programs

The waste form produced in the DWPF must be compatible with the transportation systems developed for shipping the canisters of waste to a repository. The waste forms will be the innermost of the waste package components to be emplaced in a geologic repository. The National Waste Terminal Storage (NWTS) Program has the responsibility for developing the technology and the repositories for disposal of high-level waste. Figure 2.1 of the DWPF EIS[1] shows the coordination of the DWPF with the transportation and repository programs. Information on waste form descriptions, waste package designs, product and performance specifications, repository designs, conditions, and risk analyses is routinely exchanged among all programs to ensure consistency and compatibility.

2.3.3 International Waste Form Programs

All other nations now performing or planning nuclear fuel reprocessing have selected borosilicate glass as the waste form to immobilize high-level waste (the USSR is still using phosphate glass as well as borosilicate glass). France has been in full production of vitrified (borosilicate glass) waste canisters since 1978. Belgium, Germany, Japan, the Netherlands, Sweden, and Switzerland have contracted to have their spent fuel reprocessed in France and to have the vitrified waste returned along with purified products. Current research in the European countries and Japan is focused on the development of borosilicate glass processes for immobilizing commercial high-level waste. The goal in several countries (e.g., Belgium, Germany, U. K., and Japan) is to have their own fuel reprocessing and waste vitrification facilities. Further details on these international waste form programs are given in Appendix A.

Some work is also being performed abroad on crystalline ceramic waste forms, particularly in Australia and Japan. The Synroc concept of a titanate mineral waste form, which is the basis for much of the current effort on crystalline ceramic waste forms, was originated by Professor A. E. Ringwood[8] at the Australian National University. Both Professor Ringwood and the Australian Atomic Energy Commission are continuing to develop these forms.

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3. PROPOSED ACTION AND ENVIRONMENTAL IMPACTS

3.1 PROPOSED ACTION

The proposed action is to select borosilicate glass as the waste form for immobilizing SRP high-level radioactive waste in the DWPF. Borosilicate glass was utilized as the reference waste form in the DWPF EIS.[1] The environmental consequences of selecting borosilicate glass are within the envelope of effects discussed in the DWPF and disposal system EISs.[2] The assessment also shows that the environmental effects of disposing of SRP high-level waste as a crystalline ceramic form would not differ significantly from the projected effects for disposal of the borosilicate glass form.

3.2 PROPOSED WASTE FORM

The proposed waste form for immobilization of SRP high-level radioactive waste is borosilicate glass. In the glass-making process, the high activity fraction of this waste is mixed with glass-forming chemicals and melted at 1150°C. Tests on glass made with actual and simulated waste on a small scale, and glass made with simulated waste on a large scale, indicate that borosilicate glass can accommodate different SRP waste compositions and provide acceptable levels of the following attributes:

- Waste loading
- Leach rate
- Thermal stability
- Resistance to radiation effects
- Impact resistance.

3.2.1 Description of Borosilicate Glass Waste Form

Borosilicate glass is an amorphous material formed by melting SiO_2 together with the oxides of elements such as sodium and boron. Borosilicate glass was chosen as the proposed waste form for SRP waste from among other glasses because it combines a relatively low melting temperature, 1050 to 1150°C, and high waste solubility with acceptable leach resistance and thermal and radiation stability.[3] Because of its amorphous nature, borosilicate glass can accommodate a wide range of waste compositions while maintaining favorable product and processing characteristics.

Aluminosilicate glasses have been proposed as an alternative to borosilicate glasses. However, the melting temperature of typical aluminosilicate glass is approximately 1400°C compared to the melting temperature of 1150°C for borosilicate glass. A higher melting temperature would require more development of electrode materials and ceramic refractories and would probably result in decreased melter life. Also, off-gas problems from the melter would be appreciably increased. Since the aluminosilicate glasses offer little if any improvement in chemical durability over the borosilicate glasses, it was judged that they did not justify the increased processing problems and expense.

The borosilicate glass waste form to be produced in the DWPF will consist of about 46 wt % SiO_2, 11 wt % B_2O_3, 20 wt % alkali oxides, and 23 wt % other components. This includes a waste loading of about 28 wt % (primarily oxides of iron, silicon, aluminum, manganese, and uranium). A typical composition of the glass waste form is given in Table 3-1.[4]

TABLE 3-1

Typical Composition of SRP Waste Glass

Component	Concentration, wt % Waste Glass	Contribution From Waste
SiO_2	46.3	4.8
Fe_2O_3	5.9	5.9
Fe_3O_4	2.8	2.8
Na_2O	16.3	3.8
B_2O_3	10.9	-
Li_2O	4.2	-
MnO_2	1.6	1.6
Al_2O_3	3.2	3.2
NiO	0.6	0.6
MgO	1.6	0.2
U_3O_8	1.2	1.2
CaO	1.0	1.0
TiO_2	0.7	-
ZrO_2	0.4	-
La_2O_3	0.4	-
Other solids*	2.9	2.9
	100	28

* "Other solids" include zeolite, undissolved salts, and radionuclides. Chemically, radionuclides are less than 0.1% of the waste.

The borosilicate glass waste form is made by melting a mixture of glass frit (i.e., glass former) with a wet slurry of waste in a joule-heated melter.* The molten glass is poured into canisters, 0.61 m in diameter by 3.0 m long, each containing approximately 1480 kg of glass waste. Characteristics of the reference glass canister are given in Table 3-2.[5]

TABLE 3-2

Characteristics of Reference Borosilicate Glass Waste Canister

Characteristic	Reference Borosilicate Glass[5]
Waste loading, wt %	28
Waste form weight per canister, kg	1480
Total weight of waste canister, kg	1930
Waste form density, g/cm^3	2.75
Canister material	304L stainless steel
Canister dimensions	0.61 m in diameter 3.0 m in length 0.95-cm wall
Heat generation, W/Canister (5-yr-old sludge plus 15-yr-old supernate)	423
Heat generation after 1000 years, W/Canister	<1
Radionuclide content, Ci/canister (5-yr-old sludge plus 15-yr-old supernate)	150,000
Radiation, R/hr at 1 m	2900

Borosilicate glass has been studied for the immobilization of SRP high-level waste since 1974 (Appendix B). Initial development was directed toward demonstrating the feasibility of vitrifying SRP waste through laboratory-scale tests with simulated and actual SRP wastes.[3,6] Several glass-former compositions (frits) were investigated to improve both processing and product performance

* Heating is supplied by passing alternating current through opposing pairs of electrodes positioned in the molten glass.

characteristics. In 1977, large-scale vitrification tests began with simulated SRP waste.[7] As a result of these large- and small-scale tests, glass frit compositions have been systematically improved, leading to the current frit composition, Frit 131.[8]

The properties of the borosilicate glass waste form are primarily determined by five of the glass components: silica, alkali (Na_2O and Li_2O), boron, alumina, and iron oxide. The alumina and iron oxide come from the waste itself and are particularly important determinants of the durability (mechanical stability and resistance to leaching by groundwater) of SRP waste glass.

3.2.2 Waste Form Properties

In the following sections, leach resistance, important physical properties relating to mechanical and thermal stability, and radiation stability of borosilicate glass are discussed.

3.2.2.1 Leaching Properties

Leachability is a very important property for evaluating waste forms.[9] In a multi-barrier geologic waste repository, interaction of the waste form with groundwater is the most plausible means to transfer radioactive materials to man's environment, although repository sites are being selected in those formations in which water intrusion in significant quantities is unlikely.

The most important determinants of the leachability are the borosilicate glass composition, the composition of the leachant, the leachant temperature, and the duration of exposure of the borosilicate glass to aqueous attack. Leachability is less affected by the presence of other waste package components, lithostatic pressure, or hydrostatic pressure.[4] The above factors and their effects on borosilicate glass leachability are summarized in Table 3-3. Leachability of the borosilicate glass waste form is discussed in detail in Reference 4.

At temperatures in the range of those expected for leaching of SRP waste glass in a repository (25 to 55°C), steady-state leachabilities are of the order of 10^{-3} to 10^{-4} g/m^2•day. At these temperatures, leachabilities decrease from initial values of 10^{-1} to 10^{-3} g/m^2•day, depending on the radionuclide, and then gradually approach the steady-state values.[6,10,11] Steady-state leachabilities for cesium, strontium, and plutonium in glasses containing actual SRP waste are shown in Table 3-4.

TABLE 3-3

Factors Affecting Leach Resistance of Borosilicate Glass Waste Form

Factor	Effect
Waste Loading and Composition	Increasing waste loading from 28 to 35 wt % decreases leachability by about 1/2.
Leachant Composition	Leach rates for two simulated groundwaters, brine and silicate, are typically within a factor of 5.
Leachant pH	Very little effect is expected over pH range for repository groundwaters (pH 5 to pH 9).
Duration of Exposure to Groundwater	Initial leachabilities (<28 days) are 10^{-1} to 10^{-3} $g/m^2 \cdot d$; steady-state values are 10^{-3} to 10^{-4} $g/m^2 \cdot d$.
Leachant Temperature	Decrease in temperature from 90 to 40°C results in about a factor of 10 decrease in initial leachabilities, depending on species leached and glass composition.
Leachant Flow Rate	For groundwater flows expected in repositories (<1 m/yr), variation in leachability would be small.
Pressure	Increase in pressure tends to decrease leachability, but the effect is small.

TABLE 3-4

Leachability of Actual Waste Glass in Distilled Water Based on Strontium, Cesium, and Plutonium

Waste	Element	Steady-State Leachability,* $g/m^2 \cdot d$	Release Fraction Per Year**
Tank 13	Strontium	2.6×10^{-4}	1.6×10^{-6}
	Cesium	2.5×10^{-4}	1.5×10^{-6}
	Plutonium	4.6×10^{-4}	2.8×10^{-6}
Tank 16	Strontium	1.8×10^{-4}	1.1×10^{-6}
	Cesium	2.1×10^{-4}	1.3×10^{-6}
	Plutonium	2.2×10^{-4}	1.3×10^{-6}

* Room temperature; area-to-volume ratio approximately $0.1\ cm^{-1}$.

** Calculated for a full-size DWPF canister assuming a five-fold increase in release rate due to increased area from fabrication-induced fracture.

Because the SRP high-level waste varies in composition (Table C-1, Appendix C), the effects of waste composition on leachability have been determined. In general, addition of SRP waste improves the leach resistance of the glass over that of the frit alone, primarily because of its iron and aluminum content (the major components in SRP waste). Increasing waste loading from 28 wt % (the reference loading) to 35 wt % decreases leachability by about a factor of two. Radionuclide leach rates may vary by up to a factor of five from the average over the expected range of waste glass compositions.[4,11,12]

The effects of leachant composition on glass leaching have also been studied because of expected differences in the composition of groundwater from potential repositories. The tests have shown that leachants (such as deionized and distilled water) which have low pH buffering capacity are generally more aggressive (by up to a factor of 10) than simulated repository groundwaters. However, over the range of expected repository groundwater compositions (pH 5 to pH 9), variations in pH will not significantly affect leachability.[13,14] Leach rates measured in simulated brine and silicate groundwaters are typically within a factor of 5.[4,15]

As the waste form surface temperature decreases to the ambient repository temperature due to the decay of Sr-90 and Cs-137 (Figure C-1), the leachability of the glass waste form will also decrease.* Depending on the radionuclide leached, initial (short-term) leach rates decrease by about a factor of ten as temperature is decreased from 90 to 40°C.[4,6,12] Steady-state leach rates decrease by about a factor of four over the same temperature range.[4] Thus, if the waste package should fail prematurely so that leaching occurred at 80°C (the projected maximum temperature of the design basis SRP waste glass in a wet salt repository), steady-state leach rates would be about a factor of four higher than those given in Table 3-4.

In the repository, SRP waste glass would be leached in the presence of repository minerals and multibarrier components. Tests of the interactions between SRP waste glass and other possible components of a repository system demonstrate that SRP waste glass is compatible with current repository concepts.[13] In general, the leachability decreases slightly in the presence of potential repository minerals.[4] Potential canister (304L stainless steel) or overpack (Ticode 12) materials have little effect on the leachability. Potential backfill materials can have large beneficial intezactions, and materials have been identified which have beneficial effects on glass leaching.[4]

Early results from a study of leaching mechanism of borosilicate glass suggest that the observed reduction in leach rate with time results from an adherent surface layer of oxides which forms on the glass surface and which subsequently retards leaching from the waste form matrix.[4] The controlling leaching process then becomes diffusion to and through the surface layer. Solubility limits of the waste elements in the leaching environment, however, may ultimately determine the release rate from the waste form.

3.2.2.2 Physical Properties

The importance of the mechanical and thermal properties of the waste forms is discussed briefly in Appendix B. In general, the thermal and mechanical properties of borosilicate glass are expected to be more than adequate for both normal and accident conditions that might be experienced in production, interim

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* Because of the barriers provided by the waste package and the repository, groundwater is not be expected to contact the waste form for at least 1,000 years after emplacement. At this time, the temperature of the waste form would essentially be that of the ambient repository temperature.

storage, transport, or emplacement. Also, for all normal operations, the waste canister will provide the necessary structural support. Typical mechanical and thermal properties of borosilicate glass are given in Tables 3-5 and 3-6.

A particularly important characteristic is the waste form's ability to withstand impact forces without generating and dispersing a large quantity of fines. Canisters containing Savannah River glass have demonstrated the ability to survive a 9-m drop without rupture. When subjected to impacts of 10 J/cm^3 in drop tests, samples of borosilicate glass generated very small fractions of respirable particles (Table 3-5).

Except in severe accidents, the greatest stresses to the borosilicate glass waste form will probably arise from temperature changes during cooling from the melt. Both bulk and surface cracks have been observed in initial tests with full-size canisters of simulated waste glass. However, both kinds of cracking can be limited either by controlled cooling or by use of fins in the canister. Thus, the increased surface area from cracking is not expected to increase the fractional release rate from a DWPF canister by more than a factor of five (compared to the uncracked monolith).[4,17]

In the unlikely event of a high temperature excursion (such as a fire), no volatilization would occur, and the glass would devitrify only if the temperature were maintained over 500°C for extended periods of time.[18] Because leach tests have shown that the release rate of long-lived alpha-emitting radionuclides (actinides) is not affected by devitrification, a high temperature excursion would not have a significant effect on the performance of borosilicate waste glass in the repository environment.[4]

3.2.2.3 Radiation Stability

Stability against the effects of self-irradiation is an important determinant of the waste form's long-term durability in a repository. The major cause of radiation effects in waste forms is the displacement of atoms caused by alpha particles and alpha recoil resulting from the decay of the actinide elements.[15]

Extensive radiation damage studies on borosilicate glass, including doping tests with Pu-239 and Cm-244, indicate that the performance of glass in a repository should not be affected significantly by self-irradiation for periods of 10^6 years or more.[19]

TABLE 3-5

Mechanical Properties of Borosilicate Glass[4]

Property	Borosilicate Glass
Tensile Strength, MPa	57
Compressive Strength, MPa	550
Young's Modulus, GPa	67
Poisson's Ratio	0.18
Density, g/cm^3	2.75 (100°C)
Fraction of Fines Generated in Impact of 10 J/cm^3, %	0.14 to 0.18*

* Reference 16. Fraction of particles less than 10 micrometers in size.

TABLE 3-6

Thermal Properties of Borosilicate Glass[4]

Property	Borosilicate Glass
Thermal Conductivity, W/m•K	0.95 (100°C)
Heat Capacity, J/g•K	0.83 (25°C)
Thermal Diffusivity,* m^2/s	3.8×10^{-7}
Linear Thermal Expansion Coefficient, K^{-1}	10.9×10^{-6}
Softening Point, °C	502
Annealing Range, °C	450-500

* Calculated from other properties.

3.2.3 Waste Form Processing

In the DWPF reference process, the sludge fraction of the SRP high-level waste is reacted with hot caustic in the waste tanks, if desired to reduce the aluminum content in the sludge, then washed with water to remove soluble salts. The sludge slurry is then pumped to the DWPF for vitrification. A schematic diagram of the borosilicate glass vitrification process is shown in Figure 3-1.[20]

In the DWPF, the slurry is mixed with glass-forming additives (and with any radionuclides recovered from supernate processing), heated to drive off excess water, and then fed to an electric-conduction heated, ceramic-lined melter operated at 1150°C. Here, the slurry will dry and then form a molten glass, which will be poured into a canister. After cooling to ambient temperatures, the canister will be decontaminated, sealed by welding, and then stored onsite until shipped to a federal repository for disposal.

3.2.4 Development Requirements and Goals

The vitrification process has been demonstrated on a small scale with actual waste and on a large scale with simulated waste. Each of the other key steps in the overall reference immobilization process has also been demonstrated. Laboratory tests with both simulated and actual waste have demonstrated that a durable glass waste form can be produced for SRP waste.

Optimization studies are continuing in the following areas:

- Increased solids content of melter feed slurries. Increasing the solids content from 40 to 50 wt % nearly doubled melter throughput and increased process reliability in laboratory tests.

- Increased waste content in glass. The feasibility of increasing the waste content in glass from 28 to about 35 wt % waste oxides has been demonstrated. This increase would reduce the required number of canisters at the DWPF, transportation costs, and overpack and emplacement costs at the repository, as well as improving the form's leach resistance.

- Improved glass compositions. New glass compositions have been developed which should improve melter operation and waste form performance. In laboratory tests with these glasses, corrosion of melter materials and glass volatility were reduced, compared to the reference composition. Improved frit compositions also resulted in a decrease in leachability by up to a factor of 15 (compared to the reference composition).

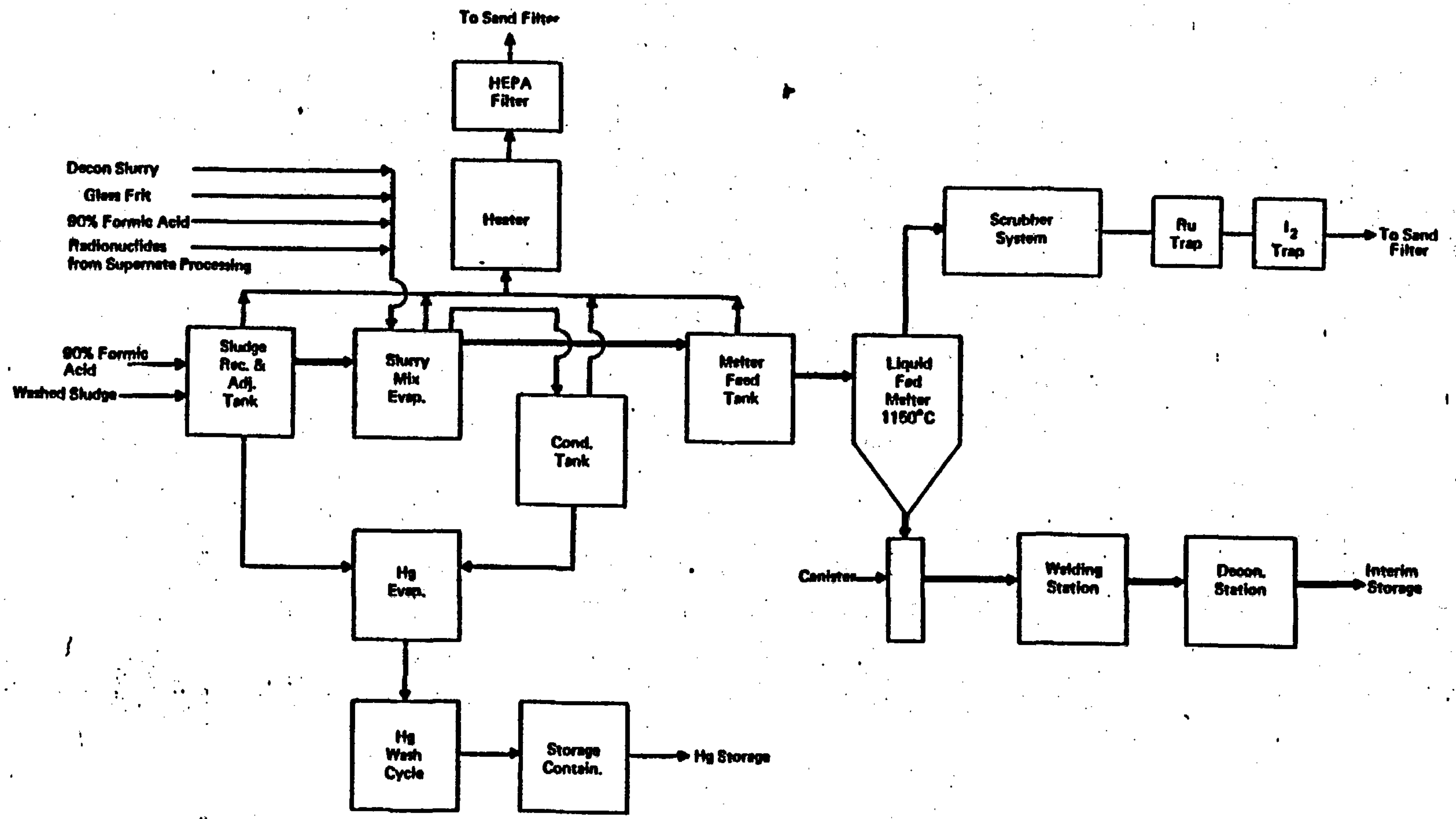

FIGURE 3-1. Borosilicate Glass Process Flowsheet[20]

- Minimizing thermal fracture in glass waste forms. Small-scale tests indicate that glass fracture during cooling from the melt can be reduced by controlled cooling and by preventing the molten glass from wetting the canister wall.[4]

- Improved repository system materials. Small-scale tests have identified promising repository backfill and other materials which reduce leach rates by up to a factor of 80.

3.2.5 Regulations and Criteria

The DWPF will be operated in conformance with all applicable Environmental Protection Agency (EPA) and DOE radiation guides for both onsite workers and the offsite public. Permits and approvals needed for the production of borosilicate glass in the DWPF were summarized in Table 6.1 of the DWPF EIS.[1]

The DWPF waste form will be shipped to a federal repository in a package that complies with applicable transportation regulations. These regulations and the responsible federal agencies are addressed in Appendix D of the DWPF EIS.

Proposed criteria and regulations that apply to federal repositories are being developed by the EPA and the Nuclear Regulatory Commission (NRC). The NWTS Program of DOE is responsible for repository operations and has proposed draft product specifications on the waste form to aid in ensuring satisfactory performance in the repository. Compliance with these repository requirements is summarized in the following sections.[21]

3.2.5.1 EPA Criteria

Although the EPA has not yet published environmental standards for high-level waste disposal, EPA has developed many internal working drafts of these criteria. The current version of the draft rule, 40 CFR 191, consists of two parts: Subpart A specifies standards for management of high-level waste and would be applicable to DWPF operations, and Subpart B contains standards for disposal and would be applicable to repository operations and closure.

Based on the latest internal draft EPA regulations, the selection of borosilicate glass as the DWPF waste form would contribute to the overall disposal system's conformance with the draft standards for management in Subpart A.

The draft criteria relating to disposal of high-level waste (Subpart B) contain projected performance requirements for repository operations in terms of total curies released to the accessible environment over a 10,000-year period. The risk assessments for typical repositories given in Section 3-4 show that virtually no activity is released in the 10,000-year period covered by the EPA criteria.

Although the number of health effects (or premature deaths) was not used as a numerical standard in the draft criteria, EPA did state that a "projected release could reasonably be limited to a level that would correspond to 1000 premature deaths over 10,000 years for a 100,000 MTHM* repository." Because the full SRP waste inventory represents an equivalent 3200 MTHM, any comparison to the EPA value for premature deaths should show that the risk is equal to or less than 32 premature deaths (10 premature deaths per 1000 MTHM). Risk analyses performed for SRP waste in a salt repository (Section 3-4) show that the dose to the affected population integrated over 10,000 years following disposal would not cause any deaths in the "best estimate" case. For an extreme case of adverse repository conditions, approximately 0.000026 premature death is estimated to occur. This is about 1 million times less than the EPA value. Under these same adverse conditions, population dose integrated over one million years is equivalent to, at most, one additional cancer.

3.2.5.2 NRC Regulations

While the NRC has no jurisdiction over defense nuclear facilities such as the DWPF, the Energy Reorganization Act of 1974 provides the NRC with specific licensing and regulatory authority over DOE facilities used primarily for the receipt and long-term storage (disposal) of high-level waste. Proposed NRC technical criteria for regulating the disposal of high-level radioactive waste in geologic repositories (10 CFR Part 60) were published for comment on July 8, 1981 (46 Fed. Reg. 35280). Most of the criteria in the proposed draft regulations pertain to repository siting, design, construction, operation, and decommissioning; however, two sections entitled Performance Objectives (10 CFR 60.111) and Requirements for the Waste Package and Components (10 CFR 60.135) relate to the waste form itself.

One of the proposed performance objectives requires that the waste package contain the waste for at least 1,000 years. This requirement on the waste package is outside the scope of this environmental assessment, but this assessment assumes that the use of borosilicate glass would contribute to the overall waste package meeting the proposed waste form performance objectives.

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* MTHM - Metric tons of heavy metal.

Another performance objective requires that the engineered system (i.e., the waste packages and the underground facility) be designed such, that after the first 1,000 years, the release rate of any radionuclide into the geological setting be less than 10^{-5} parts per year. Borosilicate glass, as part of the multibarrier approach for the waste packages, can contribute to meeting these requirements if it has leach rates $<10^{-4}$ parts per year.[22] The projected long-term release rate for the DWPF borosilicate glass waste form is below 10^{-4} parts per year, as discussed in Section 3.4.3.3.

The draft regulation on waste package requirements (10 CFR 60.135) directly includes some requirements on the waste form: the waste form must be solid, consolidated (nondispersible), and noncombustible. In addition, 10 CFR 60.135 requires that the waste package: contain no materials that are explosive, pyrophoric, or chemically reactive; contain no free liquids; be designed to contain the wastes during transportation, emplacement and retrieval; and be uniquely identified. These requirements are compatible with borosilicate glass.

3.2.5.3 DOE Specifications

The NWTS Program is developing waste form performance criteria which will include performance specifications and data requirements for high-level waste forms for geologic isolation. These performance criteria reflect all currently proposed EPA and NRC criteria that are pertinent to geologic isolation. The NWTS program has recently proposed a corresponding set of interim product specifications that include five categories of requirements (operational safety, release rate by leaching, criticality, identification, and performance testing) in three time periods:

- Operational Period (100 years after fabrication)
- Containment Period (next 1000 years)
- Isolation Period (succeeding 10,000 years).

Borosilicate glass meets the NWTS Program specifications, as described in the following paragraphs.

Operational Period. Potential safety hazards during the operational period involve damage to the canister and waste form by dropping or other impacts, or damage by fire that would allow

radioactivity to escape. Resistance of borosilicate glass waste canisters to damage by impacts and thermal excursions was noted in Section 3.2.2.2.

Similarly, borosilicate glass meets all proposed criteria with respect to combustibility, pyrophoricity, explosive properties, toxicity, and criticality.

Finally, specifications related to identification of canisters, conservatism of models used to predict long-term performance, characterization test data, and quality assurance programs can be satisfied by borosilicate glass.

Containment Period. During the containment period when heat is being generated in significant amounts by radioactive decay, it is assumed that a corrosion-resistant overpack will prevent groundwater from contacting the immobilized waste. Thus, radioactive release from the waste package by high-temperature leaching will not occur. It was earlier noted that the DWPF waste package will not, in fact, exceed 80°C at a waste surface exposed to leaching in a salt repository.

For the SRP defense high-level waste, which is characterized by low heat generation and radioactivity, the borosilicate glass waste form has demonstrated excellent thermal and radiation stability and is not expected to deteriorate during the 1000-year containment period. However, it is doubtful that such a containment period is necessary for SRP waste canisters.

Isolation Period. The waste form characteristic that is most important during the isolation period is the radionuclide release rate due to leaching, which has been tentatively specified by the NWTS Program to be less than 10^{-4} parts per year.[22] The position taken by the NWTS Program is that this release rate should be met under a variety of repository conditions to satisfy the proposed NRC criteria.

Information presently available from leach tests under simulated repository conditions indicates that the borosilicate glass waste form will meet long-term release rates of less than 10^{-4} parts per year.

3.3 AFFECTED ENVIRONMENT

3.3.1 Defense Waste Processing Facility (DWPF)

The Savannah River Plant occupies an approximately circular area of 78,000 hectares (192,000 acres) in South Carolina, 37 km southeast of Augusta, GA. The site borders the Savannah River, which forms the South Carolina-Georgia border, for about 27 km. The plant site (Figure 3-2), the DWPF site (Figure 3-3), and their environmental characteristics are described in Reference 1.

3.3.2 Transportation

The environment affected by shipping SRP high-level waste canisters is also described in the DWPF EIS.[1]

3.3.3 Generic Geologic Repository

The DOE program for isolating high-level waste emphasizes disposal in mined repositories located in stable geologic formations 600 to 1200 meters below the earth's surface.[23] The goal is to find sites in suitable rock formations that meet environmental, regulatory, and institutional requirements. Screening will identify potential sites, which will then be characterized to assess the sites' suitability for a repository. Characterization includes surface studies, boreholes to repository depth, and finally exploratory shafts.

The geologic waste repositories will be the subject of separate NEPA documentation. Appendix D gives a generic description of the repositories as a basis for determining the conditions to which the waste form will be exposed during geologic disposal, and for estimating the potential environmental consequences of repository operations and closure.

The repository site performance criteria include topics such as site geometry, geohydrology, geochemistry, geologic characteristics, tectonic environment, surface characteristics, environmental characteristics, and socioeconomic conditions.[24] Site performance and repository design features will be emphasized to ensure containment, and to provide natural and man-made barriers to waste movement. Waste migration will be further impeded by placing the repository where there are low rates of groundwater flow.[25]

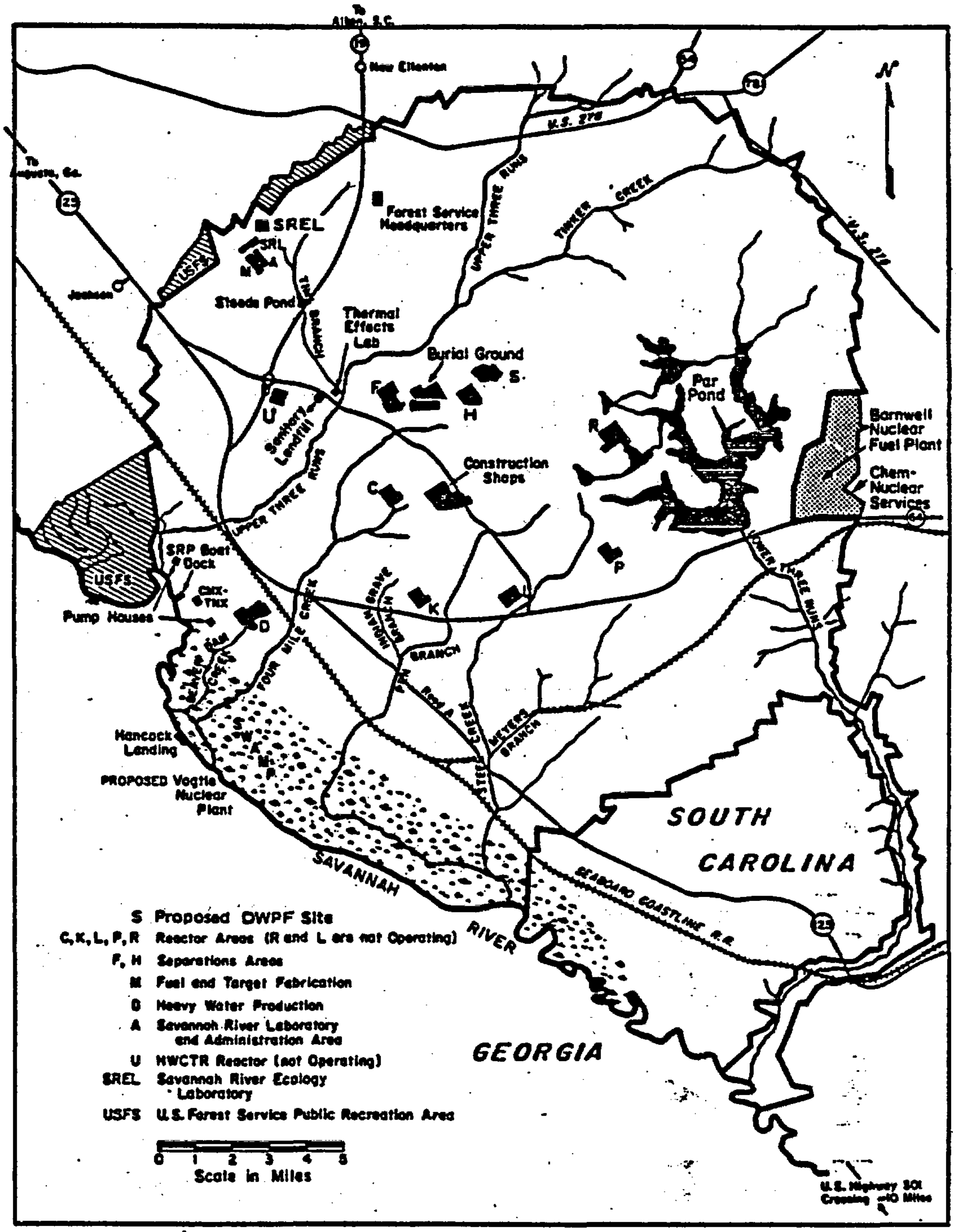

FIGURE 3-2. The Savannah River Plant Site

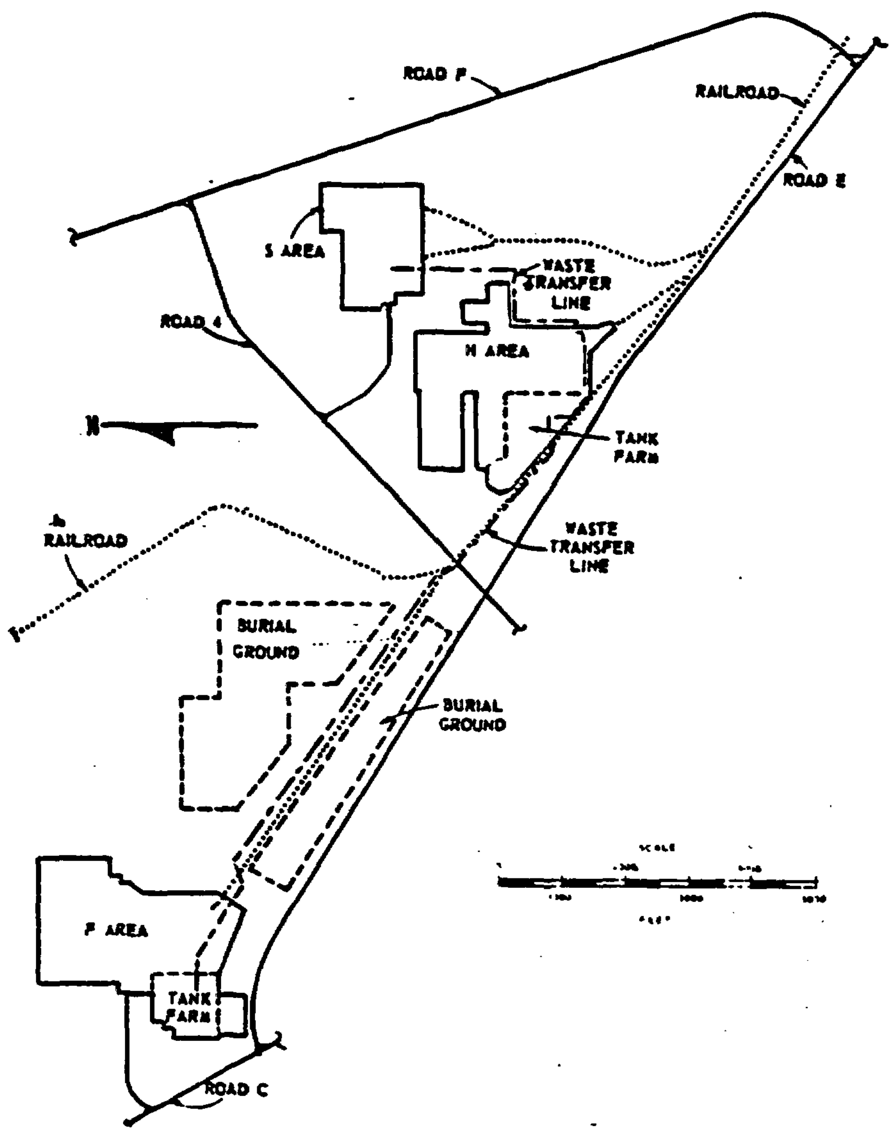

FIGURE 3-3. Proposed Location of the DWPF in S Area at the Savannah River Plant

3.4 ENVIRONMENTAL CONSEQUENCES

3.4.1 Preparation, Interim Storage, and Transportation of Borosilicate Glass Waste Canisters to Repository

The environmental impacts of immobilizing the SRP high-level radioactive waste in a borosilicate glass waste form, storing the immobilized waste at SRP until a geologic repository becomes available, and transporting the waste to a geologic repository are assessed in Reference 1. Socioeconomic effects and resource consumption from immobilization operations are minimal, and radiological effects to the public are projected to be much below normal background levels. Nonradiological effects from transportation are anticipated to be similar to those experienced with conventional common carriers. All operations will be within regulatory limits.

3.4.2 Repository Operations

3.4.2.1 Overpacking*

At the repository site, plans are for each canister of immobilized high-level waste to be sealed in an overpack designed to prevent leakage for 1000 years after the repository is closed. The overpacking will involve transferring the canister from the transport cask, handling during lag storage, placing the waste canister into the overpack, and sealing the overpack by welding.[26]

The greatest risk during the overpacking operation would be the accidental dropping of a canister onto an unyielding surface, causing breaching of the canister. Proposed DOE product specifications require the waste canister to survive a 9-m drop test (over twice the height to which a canister normally would be raised during handling) without breaching. With the proposed overpacking, the canister would be additionally protected, for example, by a carbon steel reinforcement can and by an outer titanium can. (A canister containing borosilicate glass has already passed the proposed drop test.)

* Such overpacking is a proposed requirement by the Nuclear Regulatory Commission draft of 10 CFR 60. It is designed to protect waste from contact with groundwater during an initial heat pulse period. Since the heat output of the SRP high-level waste is too low to produce a significant heat pulse, overpacking the DWPF canister may not be necessary.

The overpacking operation is performed in a conventional hot-cell in which the ventilation pattern is controlled, and all exhausts are passed through prefilters and then HEPA filters before being released to the atmosphere.

3.4.2.2 Emplacement

Emplacement includes loading the waste package into a shielded transfer cask, moving the cask to the waste hoist, lowering the hoist and cask about 640 m to the underground excavation, transferring the cask to an underground transporter, moving through underground corridors to the storage room, and emplacing the waste package into a hole in the floor of the storage room. The hole is backfilled with crushed host rock, and a concrete plug is placed on top to close the hole.

The descent of the shielded transfer cask in the waste hoist has potential for severe damage to the canister if the hoist should malfunction and allow the canister to fall freely. However, because of multiple safety features designed into the hoist, a 2000-ft fall of the waste hoist is estimated to have a probability of about 10^{-5} per year. If the fall were sufficient to breach the canister, impact tests on the borosilicate glass waste form show that less than 0.2% respirable fines would be produced in such an impact.[16]

To result in any harm to the public, hoist failure must coincide with failure of the underground ventilation system. This system is one of the major engineering features in the repository, and includes roughing filters, HEPA filters, water sprays, demisters, and multiple fans. Underground ventilation would be diverted through the multiple exhaust filter arrangement only in the event of a release of radioactivity. The probability of failure of exhaust filters is estimated to be 10^{-4} per year. The combined probability of a hoist failure and a simultaneous filter failure is 10^{-9} per year.[27]

All other operations would limit the free fall to 1.2 times the canister length (about 4 m), and are covered by the existing specification that the canister must survive a 9-m drop test without breaching. In current plans, the canister would, in fact, be doubly encapsulated in the overpack during the entire emplacement sequence.

3.4.2.3 Retrieval

Should retrieval of the waste be required after emplacement, it is assumed that only the waste canister could be retrieved because the overpack assembly would most likely be bound in the

burial hole (e.g., due to creep of salt). The retrieval scenario further assumes that the emplacement room and access corridors have been backfilled, but that the repository is still accessible.

The processes associated with retrieval of the waste package include the following:

- Location of emplacement tunnel (if sealed)
- Re-excavation of emplacement tunnel (if backfilled)
- Location of waste package (determine verticality)
- Overcoring to expose top surface of containerized waste package
- Cutting overpack and removing the overpack head pieces
- Extracting waste canister into shielded transfer cask.

After the canister is raised into the transfer cask, the cask would be moved to the main hoist and brought to the surface. At the surface, the canistered waste form would be placed in shielded storage for further disposition. The canistered borosilicate glass has the required mechanical strength to survive such an operation.

3.4.3 Long-Term Effects of Isolation

A geologic repository will be designed to control long-term radionuclide releases to levels that conform with applicable requirements. Consequence analyses of the of high-level waste disposal in geologic repositories generally conclude that the isolating qualities of the geologic media will dominate the performance of the disposal system.[28,29,30]

Once the waste is placed in a repository, natural processes over the geologic time frame could allow groundwaters to enter the repository, corrode the canister, contact the waste form, and cause the leaching of radionuclides. Contaminated groundwater would then migrate to the accessible environment (surface or underground water supplies that are used by humans). Studies of repository performance conclude that this process would be the only major contributor to the risk of human exposure.[30] Any doses to humans would occur at least thousands, and as much as millions, of years after repository closure because long periods of time would be required for the waste to leach and for the contaminated groundwater to traverse the distance between the repository and the accessible environment. Also, radionuclide travel in the groundwater generally would be retarded by sorption in the geologic media.

As a result of these time delays, which allow most of the radionuclides to decay, and the large volumetric dilution that would occur during transport, calculated doses are insignificant when compared with the effects of other natural toxic substances in the earth's crust.[31] They are also small when compared with the exposure to man from natural radioactive sources.[32,33]

3.4.3.1 Repository System Performance Models

Over geologic time periods (~10^6 years), the release of radionuclides from the repository will be governed primarily by barriers formed by the surrounding geologic media, and then by the waste form and by the engineered barriers. Geochemistry of the potential repository media is reasonably well known, and this information can be used to predict the long-term behavior of the disposed waste. As discussed in Section 3.4.3.3, migration of the radioactive components is expected to be retarded by the solubility limits of the dissolved waste and by chemical interactions (such as sorption) with the engineered barriers and the repository rock.

Several studies have analyzed the long-term performance of geologic waste isolation systems.[28-30,34-37] Typically, these studies use mathematical models to simulate and assess the behavior of the waste form, the repository site, and the overlying rock in pathways along which radionuclides could be transported to the human environment.* Values and ranges for geologic and waste form properties determined from geologic exploration and laboratory tests are used to represent interactions between the waste elements and components of the isolation system. Although the details of the analyses may differ, these studies have generally concluded that the exposure to future generations from isolated high-level wastes will be very small and that the doses will be controlled primarily by the geologic media and less so by the engineered barriers of the repository.

A typical model of the waste form/repository/site system is illustrated in Figure 3-4. Such models can be divided into three major subsystems:

- Release rates of radionuclides from the waste form and repository.

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* Several of these studies for commercial high-level waste and spent fuel are reviewed and compared in Reference 30.

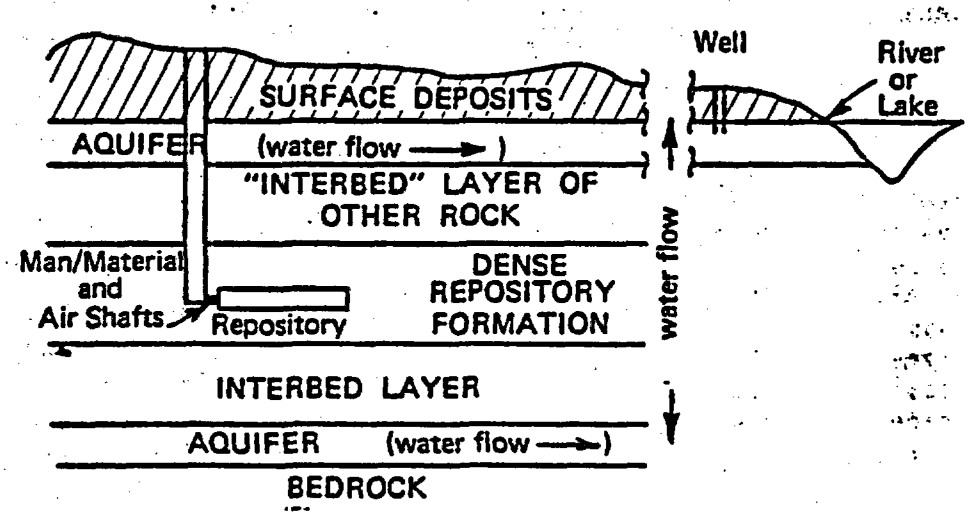

FIGURE 3-4. Typical Repository Site Model

- Hydrologic transport of radionuclides through the rock formations to a freshwater aquifer.

- Transport to and uptake by humans. Dose models are based on human-use patterns for surface water bodies (lakes and rivers) or wells drilled into an aquifer.

Several approaches have been used in evaluating the above processes which might lead to human exposure. "Deterministic" analyses choose specific values for the parameters and calculate the performance of a defined system. "Sensitivity" analyses identify which components have the most influence on the performance of the isolation system. "Uncertainty" analyses recognize that no repository can be modeled exactly; properties can be estimated only within an approximate range of values. Rather than select the "worst" possible value for each property, the analyses can treat all of the uncertainties simultaneously by a "Monte Carlo" technique. The result is a probability distribution of doses for the modeled system.

Although repository design, operations, and closure will be conducted to minimize detrimental effects on the surrounding rock, the geologic media will not be returned to their exact original state.[38] Assessments of long-term isolation, therefore, must also consider the possibility that engineered and natural barriers could deteriorate.

3.4.3.2 Performance Assessment for SRP Waste

An assessment of dose-to-man was performed for SRP waste in potential geologic repositories by Lawrence Livermore National Laboratory (LLNL).[34,35,39] This assessment included uncertainty and sensitivity analyses for undamaged ("uneventful") repositories, as well as analyses of the consequences of events which might disrupt the repository and surrounding geologic media.

Results of these analyses indicate that, under most circumstances, peak doses from SRP waste disposal will be much less than 1% of the dose from natural background radiation. Also, predicted health effects are many orders of magnitude lower than those caused by other sources. For a typical repository, credible events which might damage the repository would not significantly affect human exposure. Waste form release rates generally affect expected peak doses only if the doses are already negligibly small. For a "poor" repository site, which could yield higher, but still low doses, the waste form had little effect. These general results have recently been corroborated by an analysis which used the repository performance assessment model developed by Pacific Northwest Laboratory (PNL) for spent fuel disposal.[28,29]

Bedded Salt. Using uncertainty analyses, LLNL performed extensive studies of dose-to-man from SRP waste in a bedded salt repository.[34,35,39] Water from a lower aquifer (Figure 3-4) was conservatively assumed to permeate the salt layer to initiate the release of radionuclides from the waste. The radionuclide-containing water was then assumed to rise to an upper aquifer, from which it might be extracted by a well or might eventually contaminate surface water. Results of these processes are summarized in Table 3-7, in terms of the "best estimate" and "90% confidence level" doses for three cases:* (1) peak dose to an individual drawing all his drinking water from a well located 1.6 km downgradient from the repository; (2) peak dose to the average individual in a population residing in a river system that is fed by the upper aquifer 20 km from the repository; (3) total dose to the river system population over periods of 10^4, 10^5, and 10^6 years after repository closure.

The waste form's effect on repository system performance was assessed by assuming a mean fractional release rate of 5×10^{-6} parts per year from a waste package in salt, and associated standard deviations of one and two orders of magnitude. For the more extreme cases in the uncertainty analyses, the package release rates were generally higher than the mean. As discussed in the next section, the quoted release rate was estimated for a cracked borosilicate glass monolith, based on laboratory leach tests, making the highly conservative assumption that dissolution is not limited by solubility or by interaction with other package materials and/or rock.

5×10^{-8}
5×10^{-8}

The sensitivity of population dose and potential health effects to the release rate of the waste package is shown in Figure 3-5.[34] Dose is relatively insensitive to release rates greater than about 10^{-6}/yr for the least optimistic choices of geologic parameters (the 90% confidence level). For the "best estimate" case, doses vary appreciably with release rate less than $\sim 10^{-5}$ parts per year; however, these doses are already extremely small. Therefore, the properties of the repository site will dominate over waste form leach resistance in determining dose-to-man.

* Results of uncertainty analyses show the relative likelihood of possible doses or health effects for the parameter ranges used in the model. For example, the 90% confidence level dose is the dose that equals or exceeds 90% of the doses that are calculated by varying parameters over their possible ranges. The best estimate value represents the dose for which there are equal probabilities that doses would be greater or smaller.

TABLE 3-7

Dose-to-Man from SRP Waste in a Bedded Salt Repository

	Dose from Repository		Dose From Natural Background Radiation
	Best Estimate	90% Confidence Level	
Peak dose to a maximum individual, 1.6-km well, rem/yr	6×10^{-5}	1×10^{-2}	1×10^{-1}
Peak dose to an average individual, river system,* rem/yr	3×10^{-9}	2×10^{-7}	1×10^{-1}
Total population dose, river system,* person-rem			
10^4 yr	$<2 \times 10^{-8}$	2×10^{-1}	1×10^{8}**
10^5 yr	9×10^{0}	9×10^{2}	1×10^{9}**
10^6 yr	2×10^{2}	2×10^{3}	1×10^{10}**

* River system fed by aquifer 20 km from repository.

** Assumes a constant population of 100,000 people.

The best estimate of peak dose to the well user is about three orders of magnitude below background radiation. Even this small dose is believed to be pessimistic because of the conservatively high estimate used for the release rate. The population dose integrated over one million years is equivalent to less than one excess cancer, even at the 90% confidence level. In contrast, for a population of 100,000, more than 180 people per year would die from cancer from all causes, based on 1978 data for cancer incidence in the U.S. This would amount to about 1.8×10^8 cancer deaths over one million years compared to less than one potential death caused by the geologic isolation of SRP waste.

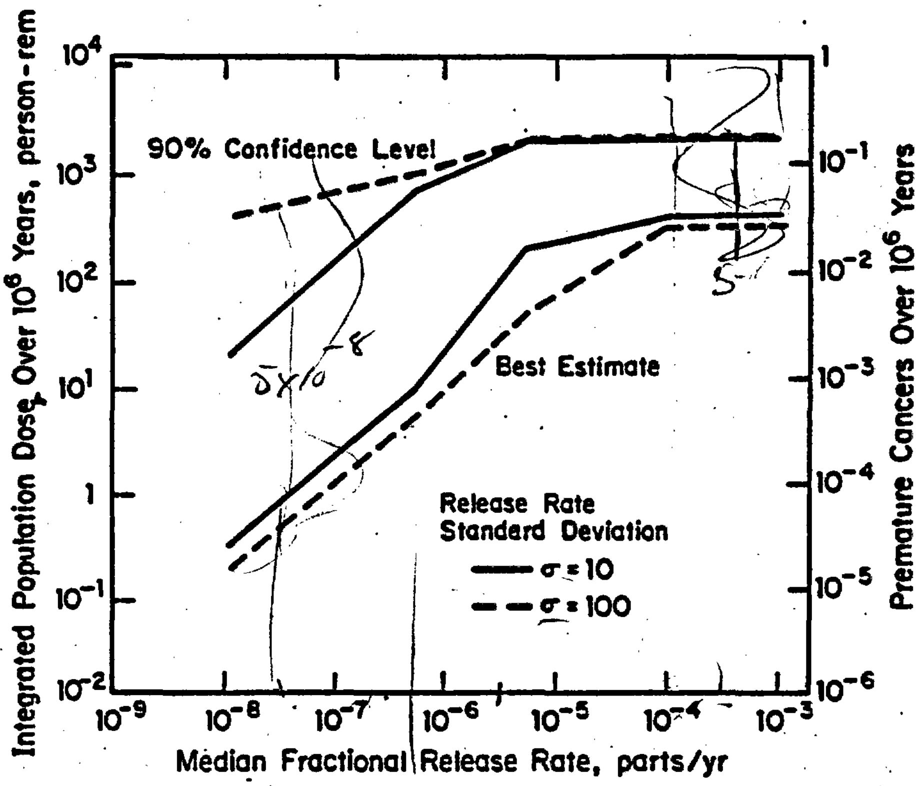

FIGURE 3-5. Sensitivity of Population Dose and Health Effects to Waste Package Release Rates

LLNL also modeled flaws and "disruptive" events, which could damage the integrity of the repository.[34] "Best estimate" doses for these cases, which include an undiscovered borehole into the repository and fault movement, are summarized in Table 3-8.

TABLE 3-8

Dose-to-Man from SRP Waste in a Disturbed Salt Repository

	Peak Individual Dose, 1.6-km Well, rem/yr	Total Population Dose Over 10^6 yr,* person-rem
Uneventful	6×10^{-5}	2×10^{2}
Fault through repository	6×10^{-3}	2×10^{3}
Failed or undetected borehole	5×10^{-3}	1×10^{3}
Deteriorated backfill	6×10^{-4}	1×10^{3}
Breccia pipe	3×10^{-4}	3×10^{2}
Dose from background radiation	1×10^{-1}	1×10^{10}**

* Based on river system fed by aquifer 20 km downgradient from repository.

** Assumes constant population of 100,000 people.

These flaws rarely increase the expected dose by more than an order of magnitude. For the 90% confidence level and higher, dose commitments actually decrease for some disruptive events.[34] Groundwater, which could pass through the entire area of an "uneventful" repository, is instead channeled along the more-permeable flows. Thus flow of water could bypass all or part of the waste in the repository.

For the disturbed salt site, reducing the waste form release rate by an order of magnitude always gave less than a ten-fold reduction in dose.

For the most severe cases modeled, LLNL showed that simple repository design features, such as providing a permeable "bypass" for groundwater underneath the repository, could reduce the doses significantly.[40]

In another study, dose-to-man calculations for SRP waste were performed with a PNL risk analysis model used previously to analyze the storage of spent fuel in a salt repository.[28,29] Results summarized in Figure 3-6 as a function of fractional release rate and groundwater travel time, generally agree with those of the more detailed LLNL analysis. The doses are generally less than 1% of background (i.e., less than 1 mrem/yr) even for very poor repository sites (i.e., short groundwater transport times).*

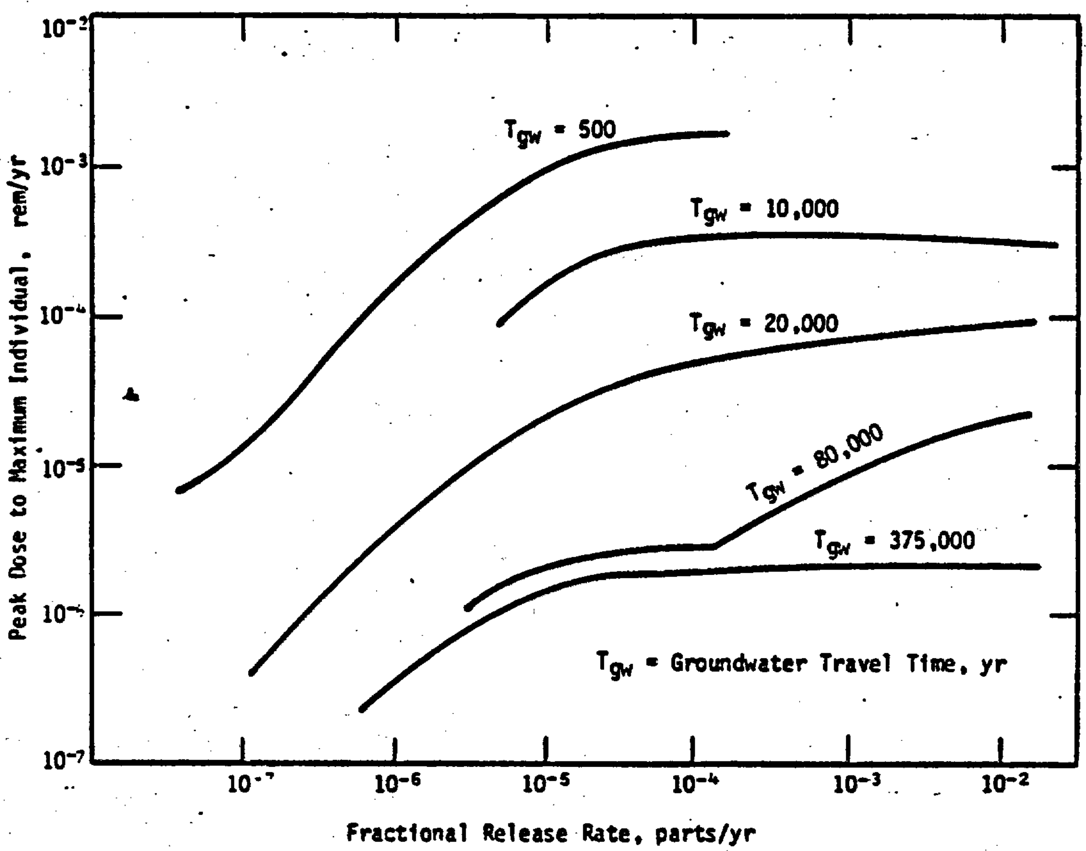

FIGURE 3-6. Dose-to-Man from SRP Waste in a Salt Repository

* The PNL study assessed the importance of groundwater travel time—the time necessary for water in an aquifer to reach a discharge point on the earth's surface. The "fractional release rate" is the rate of release into the aquifer; delays and dilution before the waste reaches the aquifer were not considered.

Basalt. LLNL also used the uncertainty analysis approach to calculate individual and population doses for SRP waste stored in a basalt repository.[34] The basalt results are summarized in Table 3-9. As in the analyses of bedded salt, maximum doses are much less than natural background.

TABLE 3-9

Dose-to-Man from SRP Waste in a Basalt Repository

		Dose from Repository		Dose From Natural Background Radiation
		Best Estimate	90% Confidence Level	
Peak dose to a maximum individual, 1.6-km well, rem/yr	Basalt	1×10^{-3}	4×10^{-2}	1×10^{-1}
	Ratio (Basalt/Salt)	15	4	—
Total population dose, over 10^6 yr,* person-rem	Basalt	1×10^{3}	2×10^{3}	1×10^{10}
	Ratio (Basalt/Salt)	5	1	—

* Based on river system fed by aquifer 20 km downgradient from repository.

The basalt doses are generally higher than the salt doses, but these differences are small at the 90% confidence level. The waste form has a somewhat smaller effect on dose for the basalt repository than for the salt repository. As for salt, the properties of the basalt repository and surrounding geologic media dominate over the waste form durability in determining dose-to-man.

Other Geologic Media. Doses have been calculated for disposal of commercial high-level waste in other geologic media considered for high-level waste disposal. Results are similar to those described above. Those studies that used pessimistic geologic and waste release parameters typically predicted doses around 1% of natural background radiation, while results of more realistic studies gave doses two to three orders of magnitude lower.[30]

3.4.3.3 Radionuclide Release Rate in Repository

The release of radionuclides from the vicinity of the waste form will be governed by the repository design and characteristics of the surrounding geologic media. Most radionculdies immobilized in the waste form have low solubilities, and their sorption on engineered barriers, such as backfill material, and on the surrounding rock should significantly reduce the release rates below those predicted from typical leach tests on the waste form.

The effects of the repository environment on waste chemistry have been considered in only a few risk studies (for example, References 36 and 37). The rate of waste release is usually treated parametrically by estimating a "release duration" over which the waste form (or repository) will release all of its contents at a constant rate.[28,29] For specific waste forms, release rates based on laboratory leach tests are generally used. However, experimental data indicate that the release of waste from the engineered system may be very much slower than the release rates based on laboratory leach tests.[41-43]

Factors affecting the release of radionuclides from the engineered barrier system of the repository include groundwater flow, oxidation-reduction conditions, temperature, pH, solubility of the leached radionuclides, and interaction of radionuclides with surrounding materials (such as sorption). The effects of these factors on the release of radionuclides from the SRP borosilicate glass waste form are discussed below.

A repository in bedded or domed salt would be expected to have no natural groundwater flows, at least for long time periods. If water penetrates a salt repository, the flows would be extremely slow and would result in essentially static leaching conditions. Crystalline rock media (such as basalt, tuff, shale, and granite) are characterized by very slow movement of underground waters, and would also provide virtually static leaching conditions. Only for unlikely geologic or man-caused events could a significant flow of water pass through the repository.[34]

Natural groundwaters contain little dissolved oxygen. Under these reducing conditions, the actinides and technetium have such low solubilities that they would not dissolve at significant concentrations.[33] Most leaching tests, however, have been performed with water in contact with air; the soluble species measured in these tests are believed to overstate the actual release of these elements in a repository which fills with groundwater after closure. Whereas salt repositories are not expected to fill with water, repositories in granite and basalt are expected to be below the water table and, after closure, will slowly fill with water. In repositories which do fill with water after closure, water could dissolve oxygen from trapped air and create oxidizing conditions.

This dissolved oxygen would soon disappear, however, because of interactions with the rock.[37,41] Thus, long-term leaching of waste forms should be under reducing conditions which would tend to limit the solubilities of the radionuclides.

After the short-lived radioactive elements have decayed, temperatures in the repository will approach the ambient temperatures of the surrounding rock. Typical ambient temperatures for salt are around 35°C;[44] hardrock conditions would range from 20°C in granite to about 50°C in basalt.[45] Leaching and other waste element interactions would be expected to occur at these temperatures.

A range of radionuclide release rates that might occur in a repository can be estimated by using laboratory leaching data to establish an upper bound, coupled with available solubility data to provide a lower, more realistic estimate for the insoluble elements. For the LLNL analyses, fractional release rates in salt (5×10^{-6} parts per year) and basalt (10^{-5} parts per year) were conservatively estimated using available leaching data on borosilicate glass,[46,47] correcting for temperature, and assuming a five-fold increase in release rate due to fabrication-induced cracking. For insoluble radionuclides, such as most of the actinides and technetium, release rates would most likely be controlled by their solubilities in the groundwater. Release rates of actinides predicted from solubilities are generally orders of magnitude lower than the rates estimated from leaching data.[36,48]

Other interactions between the waste form, groundwater, and natural and engineered barriers could also lower release rates from those estimated based on leaching tests. For example, insoluble products of leaching can create a protective layer on the waste form's surface. Such protective layers have been observed on leached surfaces of borosilicate glass.[46,49]

Surrounding rock can also contribute to the retardation of waste migration by reacting with waste species. Although not representative of expected repository conditions, high-temperature leach tests of borosilicate glass in the presence of crushed granite, basalt, or salt, showed three orders of magnitude less uranium in solution with rock present than without the rock.[42,50] Silicon, sodium, and cesium concentrations in solution were also greatly lowered.[42]

Other materials in the repository can also limit the intrusion of water and impede waste transport. Backfill clays, for example, could delay the movement of actinides from the vicinity of the waste form canister for up to 100,000 years.[51] Other materials can control groundwater chemistry or strongly sorb radionuclides.[52] In addition, the presence of certain canister materials may lower

leach rates; e.g., borosilicate glass leach rates have been observed to decrease by up to two orders of magnitude in the presence of lead.[43,46] Aluminum can also decrease leach rates.[41]

In summary, the complex interactions of the waste elements with other materials in the repository, their solubility limits, the long duration of groundwater travel, and sorption of the waste elements in the surrounding geologic media will combine to limit release of radionuclides to the accessible environment to values much lower than those estimated from simple laboratory leaching tests. In particular, the following effects are expected for some specific radionuclides:[33]

- The transport time of the most hazardous fission products, Sr-90 and Cs-137, would be long enough to permit their full decay.
- Sorption of long-lived actinides, such as americium and plutonium, would retard their movement through the geologic medium, permitting substantial decay before potential release.
- Weakly sorbed long-lived radionuclides, such as Tc-99, Np-237 and Ra-226, would be only slightly soluble in groundwaters expected in deep geologic formations. Thus, their movement with groundwater would also be retarded, and the potential hazard to humans would be reduced.

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4. ALTERNATIVE WASTE FORM (CRYSTALLINE CERAMIC)

The screening process described in Appendix B identified crystalline ceramic as the primary alternative waste form to borosilicate glass. Crystalline ceramic is a generic term for a product of compatible mineral phases, formed at high temperatures. Two candidate waste forms, Synroc-D (a titanate-based ceramic) and tailored ceramic (an alumina/rare earth-based ceramic), are included in this term. In laboratory tests with simulated waste, the ceramic form has exhibited low leach rates, especially for uranium. Its mechanical and thermophysical properties are comparable to those of borosilicate glass, and its stability to damage from self-irradiation should be adequate based on studies with natural analogues. The process for immobilizing SRP high-level radioactive waste in crystalline ceramic is feasible, but is significantly more complex than the borosilicate glass process. The calculated environmental impacts resulting from production and dispasal of the ceramic form are essentially the same as for the borosilicate glass waste form.

4.1 DESCRIPTION OF CERAMIC WASTE FORM

The crystalline ceramic waste form is a dense compact of compatible fine-grained oxide phases. Each of these phases serves as a "host" for one or more of the radioactive or inert elements present in SRP waste.[1] The ceramic form of primary interest for SRP waste immobilization is Synroc-D developed by LLNL[2] based on original work done by A. E. Ringwood at the Australian National University.[3] The expected phases in Synroc-D and the waste elements they contain are shown in Table 4-1.[1]

The Synroc-D form was designed specifically for SRP waste and utilizes titanate phases, zirconolite and perovskite, as the primary crystalline hosts for radionuclides. These phases are similar to natural minerals which have effectively retained radioactive elements for millions of years.[4] Synroc-D also includes other oxides, largely derived from the waste itself, such as spinels and nepheline, which accommodate large quantities of iron, aluminum, and sodium. The spinel phases would include essentially no radioactive elements, whereas nepheline and a related intergranular glassy phase contain cesium.[2]

TABLE 4-1

Typical Composition of Ceramic (Synroc-D) Phases with SRP Waste[1]

Mineral Phase	Approx. Phase Composition, wt %	Nominal Chemical Formula	Waste Elements*
Spinel	29	$FeAl_2O_4-Fe_2TiO_4$	Al, Fe, Mn, Ni
Perovskite	21	$CaTiO_3$	Sr, Ca, Ce, Nd, Act(III)**
Zirconolite	26	$CaZrTi_2O_7$	U, Ca, Act(IV)†
Nepheline and Glassy Si-Rich Phase	24	$NaAlSiO_4$	Na, Cs, Al, Si

* Important radionuclides are underlined.

** Trivalent actinides.

† Tetravalent actinides.

To promote the formation of these desirable phases, oxides or salts of titanium, zirconium, silicon, and calcium are added to the SRP waste feed before it is consolidated. Consolidation is accomplished at high temperatures and pressures to facilitate migration of chemical species to the favored phases and to densify the mixture. After consolidation, individual oxide grains are 1 to 2 micrometers in diameter or smaller.[2] For well-blended waste, about 65 wt %* sludge could be immobilized in Synroc-D with 35 wt % "tailoring" additives. The overall composition of Synroc-D containing well-blended SRP waste sludge is shown in Table 4-2. Unlike borosilicate glass, variations in waste composition could affect the ceramic's waste loading; for example, a large increase in Al_2O_3 content would result in a decrease in waste loading and radionuclide content.

* Without aluminum removal; waste loading on equivalent basis with borosilicate glass is ~52 wt %.

TABLE 4-2

Composition of Synroc-D and Waste Mixture Prior to Consolidation[1]

Constituent	Concentration in Mixture, wt % SRP Sludge	Additive
Fe_2O_3	18.9	—
Al_2O_3	17.9	—
MnO_2	4.3	—
U_3O_8	2.6	—
CaO	3.0	4.2
NiO	1.3	—
SiO_2	8.9	1.4
Na_2O	5.3	—
(Ca, Ba, Pb) SO_4	0.6	—
ThO_2	0.5	—
Others[a]	2.1	—
TiO_2	—	20.1
ZrO_2	—	8.8
Total	65.5	34.5

The ceramic form, as currently envisioned, would be hot isostatically pressed in a carbon steel container. The reference ceramic canister would contain three such compacts enclosed in a stainless steel canister of about the same dimensions as the reference glass canister.[1] Major features of the canistered ceramic waste form are given in Table 4-3.

4.2 WASTE FORM PROPERTIES

In the following sections, leach resistance, important physical properties relating to mechanical and thermal stability, and radiation stability are summarized for the Synroc-D waste form. These properties were measured from Synroc-D samples containing simulated (nonradioactive) SRP waste.[2,3,5]

TABLE 4-3

Characteristics of Canistered Ceramic (Synroc-D) Waste Form

Characteristic	Synroc-D Ceramic[1]
Waste loading, wt %	65*
Waste form weight per canister, kg	2400
Total weight of waste canister, kg	3650
Waste form density, g/cm^3	4.0
Canister material	304L stainless steel
Canister dimensions	0.61 m in diameter 3.0 m in length 0.95-cm wall
Heat generation, W/canister (5-yr-old sludge plus 15-yr-old supernate)	1270
Heat generation after 1000 years, W/canister	<2
Radionuclide content, Ci/canister (5-yr-old sludge plus 15-yr-old supernate)	450,000
Radiation, R/hr at 1 m	~8700

* Without aluminum removal; waste loading on equivalent basis with borosilicate glass is ~52 wt %.

4.2.1 Leaching Properties

The Synroc-D waste form is expected to be very resistant to leaching by groundwaters in geologic repositories based on early leach test results.[2,6] Leaching data available on Synroc-D are primarily from MCC leach tests* for short time intervals (28 days or less) with simulated groundwater leachants.

Synroc-D leach rates for cesium, strontium, and uranium (generated in MCC-1 static leach tests) are summarized in Table 4-4. Leach rates of the short-lived fission products—primarily Cs-137 and Sr-90—would be important for accident

* Proposed standard waste form tests developed by the Materials Characterization Center of Pacific Northwest Laboratory.[7,8]

TABLE 4-4

Cesium, Strontium, and Uranium Leach Rates for Synroc-D*

Leachant	Leach Rate, $g/m^2 \cdot day$**		
	Cesium	Strontium	Uranium
Deionized Water	0.80	0.33	0.00008
Silicate Water	0.38	0.09	0.00028
Brine	<0.37	<0.10	0.0005

* Made with composite (blended) simulated waste.

** Values listed are average 28-day leach rates at 90°C from MCC-1 tests performed by LLNL, MCC, and SRL.[6]

conditions, which would expose the waste form to water during the operational and thermal periods of waste disposal. These periods include interim storage, transportation, and the first few hundred years in the repository. Leach resistance for uranium and other long-lived actinides is of interest for the entire geologic isolation period.

Synroc-D leach rates measured in short-term MCC tests are comparable to those of borosilicate glass for cesium, are higher for strontium, and are lower for uranium.[6] Other major results of leaching studies on Synroc-D include:[2,6]

- Leaching of the multi-phase Synroc-D ceramic is incongruent (that is, varies depending on element leached) because some phases retain the waste elements more strongly than other phases; for example, zirconolite retains uranium more effectively than nepheline and the intergranular glassy phase retain cesium.

- The effects of waste composition and leachant composition on leaching are relatively small; changes in leach rates from these effects are typically less than a factor of 5.

- The effect of flow rate is variable; however, at the lowest flow rate studied, which corresponds most closely to expected flow in a repository, leach rates are about the same as in static leach tests.

The long-term resistance to leaching of Synroc-D by groundwater is difficult to predict accurately from the short-term MCC leach tests because of the different durabilities of the Synroc phases and the lack of information on protective layer formation. Generally, the silica-rich phases (nepheline and the intergranular glassy phase), which contain cesium and some strontium, are least durable, while zirconolite (which contains uranium) is the most resistant to leaching. Release rates in a repository will depend upon interactions between the groundwater, waste form, other engineered barriers, and phases formed by precipitation of components released from the waste form.

4.2.2 Physical Properties

The Synroc-D form is a hard, high-strength ceramic with mechanical and thermophysical properties listed in Tables 4-5 and 4-6, respectively. These physical properties are, in general, similar to those of the borosilicate waste form. In particular, the quantity of respirable fines (<10-μm particles) generated in an impact test of 10 J/cm^3 energy density was only 0.16%, which is approximately equal to the fines fraction generated from borosilicate glass in similar tests.

The effects of self-irradiation over long isolation periods on the properties of the Synroc-D waste form are not as well characterized as for borosilicate glass. However, evidence from studies of natural zirconolite and perovskite phases containing uranium and thorium indicate that Synroc should remain a durable host for the actinides for at least 10^6 years of geologic isolation.[2,4] The major damage mechanism in Synroc would be atom displacement caused by alpha decay, which could produce loss of crystal structure (metamictization), volume expansion and associated cracking, and increased leachability. Natural mineral studies of zirconolite and perovskite show metamictization beginning about 10^{18} to 10^{19} α/cm^3 (projected exposure for one million years of repository storage), and volume increases of 2 to 3%, but no significant increase in uranium leach rates.[4]

4.3 CERAMIC WASTE FORM PROCESSING

A potential production process for manufacturing a ceramic waste form in the DWPF was defined in the alternative forms processability study.[10] A schematic diagram of major steps in the process is shown in Figure 4-1. This process is considerably more complex than the reference glass process (Section 3.2.3) and would require a larger and more expensive processing facility.

TABLE 4-5

Mechanical Properties of Synroc-D[2]

Property	Synroc-D
Tensile Strength, MPa	75.9*
Compressive Strength, MPa	280
Young's Modulus, GPa	139
Poisson's Ratio	0.28
Density, g/cm^3	4.0
Fraction of Fines Generated in Impact of 10 J/cm^3,** %	0.16

* For Synroc-C (Synroc formulation for simulated commercial power reactor waste).

** Reference 9. Fraction of particles less than 10 micrometers in size.

TABLE 4-6

Thermal Properties of Synroc-D[1]

Property	Synroc-D
Thermal Conductivity, W/m•K	1.85 (20°C) 1.91 (200°C)
Heat Capacity, J/g•K	0.74 (20°C)
Thermal Diffusivity,* m^2/s	6.5×10^{-7}
Linear Thermal Expansion Coefficient, K^{-1}	11×10^{-6}**
Solidus Temperature, °C	1270

* Calculated from other properties.

** For 22 to 950°C.

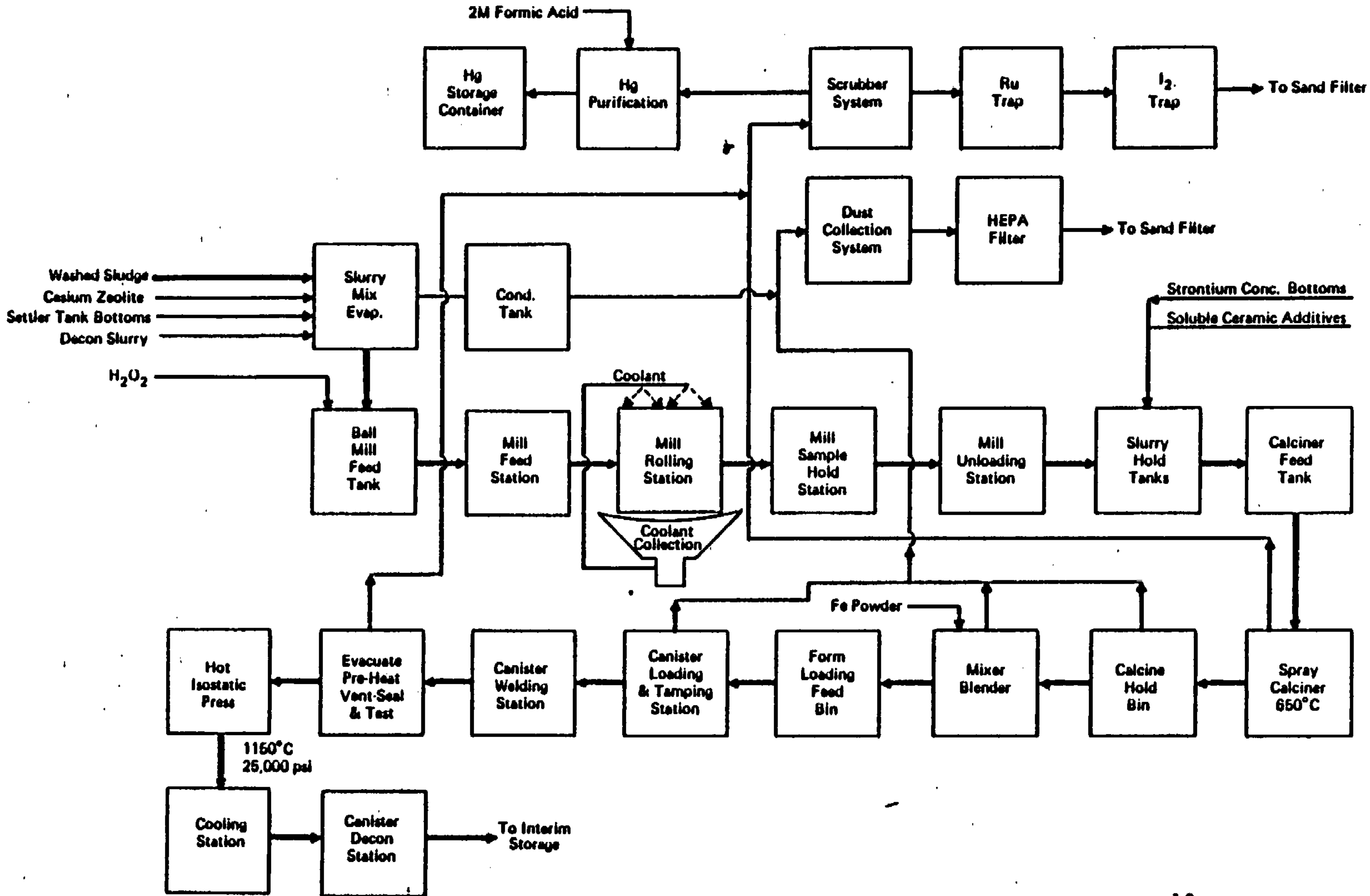

FIGURE 4-1. Crystalline Ceramics Process Flowsheet[10]

The ceramic process starts with essentially the same waste feed streams as does the reference borosilicate glass process except that aluminum is retained in the sludge feed. Washed sludge is combined with process recycle streams and cesium-loaded zeolite from supernate processing and concentrated to 40 wt % solids. The concentrated slurry is ball milled to reduce particle sizes in the feed. The milled slurry is mixed with the small amount of strontium removed from the supernate and with chemicals added to achieve the desired composition. The mixture is then spray calcined at 650°C. The calcined powder is blended with iron powder (to control cation oxidation states during consolidation), loaded into carbon steel canisters, and tamped to 50% theoretical density.

The canister is heated under vacuum to 800°C to eliminate residual volatiles, sealed, and placed in a hot isostatic press (HIP). In the HIP, the canister and its contents are isostatically pressed in argon at 170 MPa pressure and 1150°C. At this temperature and pressure, the volume of the canister decreases by 50%, and the density of the ceramic approaches the theoretical density of 4.0 g/cm^3. Formation of the desired phases occurs simultaneously with the reduction of porosity. Three carbon steel canisters, 0.56 m in diameter by 0.91 m high, are stacked inside a stainless steel canister, 0.61 m in diameter by 3.0 m high (dimensionally the same as the reference borosilicate glass canister). The waste canister is sealed by welding, decontaminated, and then transferred to an interim storage facility until a geologic repository becomes available.

4.4 DEVELOPMENT REQUIREMENTS AND GOALS

Extensive laboratory tests have been performed to develop and characterize the Synroc-D form with simulated SRP waste,[2,5] and a process for producing the ceramic has been demonstrated on a laboratory scale.[5] A potential production process has been defined, and from it a conceptual design of a ceramic waste form processing facility was developed.[10] Future development efforts would involve: (1) scale-up and demonstration of process equipment, unit operation tests, and integrated process tests; and (2) optimization of the ceramic form's phase chemistry.

Equipment development requirements identified for the ceramic process are extensive and include:[10] a vacuum ball mill suitable for remote operations, a modified remotely operated pipe connector with special provisions for evacuating and sealing containers, a sampling system for slurry particle size determination, a calciner atomization system, a monitoring system for calciner skin temperature, a fluid energy mill for calcine pickup, an in-can tamper, a remote HIP, and a canister resistant to nonuniform collapse. In

general, these needs will require invention and extensive development. Other process-related areas requiring development include process control methods and techniques to minimize dusting. Product development requirements include: hot cell testing to demonstrate that a high-quality ceramic form can be made with actual waste, and actinide doping studies to demonstrate the effects of self-irradiation on the long-term stability of Synroc-D.

Optimization studies could lead to product and process improvements in the following areas:[2,5,10,11]

- Optimizing the phase chemistry to decrease leachability of cesium and strontium from silicate phases. Both LLNL and Rockwell Science Center have shown that improvement in leach resistance of up to a factor of 10 for strontium is possible.

- Demonstrating that selectively milling only the larger particles in the sludge feed (thereby reducing the size and cost of ball milling) does not affect adversely subsequent phase formation and radwaste partitioning during consolidation.

- Optimizing the calcination step to improve reliability. Fluidized bed as well as spray calciners merit consideration.

- Optimizing the hot consolidation step to improve product quality and process flexibility.

4.5 ENVIRONMENTAL CONSEQUENCES

4.5.1 Preparation, Interim Storage, Transportation, and Repository Operations

The environmental effects of immobilizing SRP high-level waste in Synroc-D, storing the ceramic waste canisters at the DWPF until a geologic repository becomes available, transporting the waste canisters to the geologic repository, and operating the repository would be very small and similar to impacts projected for the borosilicate glass waste form (Sections 3.4.1 and 3.4.2).[12,13] Minor differences would result from a larger DWPF required for the ceramic form and from a smaller number of ceramic canisters to be shipped and emplaced in the repository, but these differences would not affect ability to operate within applicable regulations. Overall risks from release of radioactivity to the environment from extreme transportation accidents, from repository operations or from long-term isolation are proportional to the total quantity of high-level waste transported to and emplaced in the repository and would be approximately the same for the ceramic and the glass waste form.

4.5.2 Long-Term Effects of Isolation

Like borosilicate glass, Synroc-D would be a suitable waste form for long-term isolation of SRP waste. No phenomena have been observed that would significantly degrade the ceramic's ability to limit radionuclide release from a repository. Although no long-term leaching data or data for forms containing actual waste exist, MCC tests have shown uranium leach rates in particular to be very low for Synroc (Section 4.2.1). Under expected repository conditions, actinides with low solubilities might be released at an even lower rate.

As discussed in Section 3.4.3, release rates in this range would yield negligibly small doses.

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APPENDIX A

INTERNATIONAL WASTE FORM PROGRAMS

INTERNATIONAL WASTE FORM PROGRAMS

Many countries, including the United States, have been performing research and development on high-level waste immobilization for decades. France decided 20 years ago to vitrify all high-level waste generated in their nuclear power program, and to export equipment, plants, and technology. Many countries including Belgium, Germany, Japan, the Netherlands, Sweden, and Switzerland have contracted or reached agreements for France to reprocess their spent fuel and return the purified products plus a vitrified waste to them. The fact that France has a licensed and successfully operating vitrification process (AVM) weighs heavily on the selection of initial immobilization facilities in the European countries. As discussed below, many countries including Belgium, Germany, and the United Kingdom have purchased the French technology, but are also investigating other glass processes for possible later use.

Belgium. No decision has been reached as to whether the Belgian government will take over the decontaminated and decommissioned Eurochemic reprocessing plant. However, accumulated high-level waste will be vitrified in one of two facilities (high-level waste from processing highly enriched Materials Test Reactor fuels was separated from that produced from low-enriched oxide fuel and may be processed separately).

One process will be the French AVM technology (vitrification facilities at Marcoule); the other will utilize a joule-heated ceramic-lined melter designed by DWK (German fuel reprocessing company) to produce either glass beads in metal matrix (called PAMELA) or glass monoliths. Both projects are under construction and should start processing radioactive wastes in 1987 and in 1985, respectively.

France. Vitrification of high-level waste is well developed in France and still is being improved upon. PIVER, a hot pilot plant, operated from 1969 to 1973 at Marcoule, producing 12 MT of glass with batch pot calcination/melter technology. ATLAS, a half-scale prototype AVM, started up in 1978 and processed 4 m^3 of high-level waste to verify off-gas treatment requirements. A full-scale AVM rotary calciner and inductively heated melter also started up in 1978 and has processed 230 m^3 of high-level waste, yielding 108 MT of glass in 360-kg canisters (0.5 m in diameter by 1.0 m high). At the La Hague reprocessing center, three scaled-up AVM vitrification units (AVH) are being constructed and are scheduled to start up in mid-1986.

The French are currently storing the canistered waste in air-cooled vaults. Current thinking is to store the vitrified waste in surface vaults for about 50 years and then dispose of the waste in geologic repositories.

Germany. Originally, the Germans had planned a large spent fuel storage, reprocessing center, and waste disposal site at Gorleben, above salt domes proposed for geological disposal of vitrified waste. Political considerations have caused that plan to be abandoned, and now multiple strategies are being evaluated from direct disposal of spent fuel to construction of multiple, small reprocessing plants. Germany has accumulated some 65 m^3 of high-level waste at the WAK pilot plant at Karlsruhe and is currently adapting the French AVM technology to German licensing requirements for vitrifying this waste. The Karlsruhe waste facility, called HOVA, should go into hot operation by 1986.

The Germans have been actively developing a liquid-fed, joule-heated melter over the past seven years. They will test this technology at Mol, Belgium (DWK is building the melter for Eurochemic waste vitrification), and may substitute it for the AVM technology when German reprocessing plants are finally authorized.

United Kingdom. Although the British have spent many years developing rising-pot vitrification processes (FINGAL, then HARVEST), the decision has been made to go with AVM technology at Windscale. A modest development program has been started on joule-heated melters for possible use in later years.

The British, like the French, have opted for several decades of interim surface storage of the vitrified waste before transferring it to geologic disposal.

Japan. The Japanese are committed to nuclear power and, therefore, to closing the fuel cycle within Japan. Initially, however, Japanese spent fuels will be reprocessed by BNFL (U.K.) and COGEMA (France). Japan has already achieved an active reprocessing pilot plant and plans to have a commercial plant operating by 1990. Significant progress has also been made in development of high-level waste vitrification. A vitrification pilot plant is planned for 1987 to take the high-level waste from the Tokai Mura fuel reprocessing plant. To date, an engineering test facility has demonstrated operation of two types of full-scale, joule-heated melters, and a mock-up test facility and (hot) chemical processing facility are expected to start operation in mid-1982.

The Japanese, like the French and British, favor interim surface storage of the vitrified waste before ultimate geologic or seabed disposal.

APPENDIX B

WASTE FORM SCREENING

WASTE FORM SCREENING

Important Waste Form Properties

The disposal of high-level radioactive waste is planned to be achieved through the immobilized emplacement of the waste in a deep geologic repository designed to provide multiple barriers to the release of radionuclides to the environment. Current reference designs for geologic repositories include a requirement that the stabilized waste form provide one of the many barriers to the release of radionuclides. Waste form properties that contribute to this function as a barrier include:

- Low leachability — the ability of the waste form to resist chemical dissolution in natural aqueous environments. Natural groundwater could provide a means both to degrade the waste form and to transport dissolved radionuclides to humans.
- Mechanical stability — the ability of the waste form to resist mechanical dispersion and to limit the surface area exposed to leaching.
- Radiation stability — the ability of the waste form to resist chemical or structural degradation due to radioactive decay of its radionuclides.
- Thermal stability — the ability of the waste form to resist chemical and physical degradation during the period when significant decay heat is generated in the waste.

Other waste form properties or characteristics important during production, handling, interim storage, shipment, repository emplacement, and repository retrieval (if required) are:

- Processing flexibility - the process must provide a consistent quality product over a range of operating parameters.
- Waste compatibility - the waste form must be able to accommodate the expected variations in waste composition.
- Mechanical strength - the waste form must resist thermal stress and the stress of normal handling.
- Impact resistance - the waste form must minimize the quantity of dispersible or respirable particles that would be produced by an impact accident.
- Fire resistance - the waste form must not release volatile radionuclides or generate gas which might rupture the canister during accidental external fires.

Finally, other waste form attributes could impact the costs of waste form production and disposal. These include:

- Process complexity - determines capital and operating expenses for waste form production.

- Waste loading - affects the number of waste canisters to be produced, packaged, shipped, and emplaced in the repository.

Candidate High-Level Waste Forms

The evaluation of potential waste forms for immobilization of SRP high-level waste began in 1973. In 1977, borosilicate glass was selected as the reference form for the DWPF. Since 1979, seventeen candidate materials (Table B-1), including borosilicate glass, have been considered as potential solid forms for the immobilization and geologic disposal of high-level waste. Screening evaluations[1,2] during 1979 and 1980, based on performance potential and predicted process complexity of each form, reduced the number of forms from seventeen to seven. The evaluations considered nine scientific and nine engineering parameters affecting the long-term performance and production of waste forms. The elimination of ten of the forms from consideration was based upon such technical concerns as high porosities, high leach rates, questionable fracture behavior and tensile strength, incomplete partitioning of radionuclides within phases, possible effects of waste stream variation on phase assemblage and microstructure, potentially high corrosion rates, and potential phase sensitivity to radiation damage. Following continued development and characterization, the seven remaining forms (Table B-2) were evaluated further to select, in November 1981, two candidate forms for immobilizing SRP high-level waste.[3]

The selection of two of the seven forms for further development was based on four major inputs: (1) preliminary waste form evaluations conducted by the DOE defense waste sites for defense high-level waste and by an independent laboratory for commercial high-level waste; (2) peer review assessments and recommendations; (3) an evaluation of waste form product performance; and (4) an evaluation of waste form processability. The next two sections discuss the four major inputs considered in evaluating the seven candidate waste forms and the selection of the final two waste forms.

TABLE B-1

Candidate Waste Forms Considered for Geologic Disposal of High-Level Waste

Waste Form	Developer/Contractor
Borosilicate Glass	Pacific Northwest Laboratory Savannah River Laboratory
High-Silica Glass	Catholic University of America NPD Nuclear Systems, Inc.
Phosphate Glass	Pacific Northwest Laboratory Brookhaven National Laboratory
Clay Ceramic	Rockwell Hanford Operations Pacific Northwest Laboratory
Glass Ceramic	Idaho Chemical Processing Plant
Tailored Ceramic	Rockwell International Pennsylvania State University
Synroc	Lawrence Livermore National Laboratory Argonne National Laboratory North Carolina State University
Titanate Ion Exchanger	Sandia National Laboratories
Stabilized Calcine	Idaho Chemical Processing Plant
Pelletized Calcine	Idaho Chemical Processing Plant
Normal Concrete	Pennsylvania State University Savannah River Laboratory Oak Ridge National Laboratory
Hot-Pressed Concrete	Pennsylvania State University
Concrete Formed Under Elevated Temperature and Pressure (FUETAP)	Oak Ridge National Laboratory
Matrix Forms	Pacific Northwest Laboratory Argonne National Laboratory
Coated Sol-Gel Spheres	Oak Ridge National Laboratory
Cermet	Oak Ridge National Laboratory
Disc-Pelletized Coated Particles	Pacific Northwest Laboratory Battelle Columbus Laboratories

TABLE B-2

Seven Candidate Waste Forms Evaluated for Geologic Disposal of High-Level Waste

Waste Form	Developer/Contractor
Borosilicate Glass	Pacific Northwest Laboratory Savannah River Laboratory
Synroc	Lawrence Livermore National Laboratory Argonne National Laboratory North Carolina State University
Tailored Ceramic	Rockwell International Pennsylvania State University
High-Silica Glass	Catholic University of America NPD Nuclear Systems, Inc.
Concrete Formed Under Elevated Temperature and Pressure	Oak Ridge National Laboratory
Coated Sol-Gel Spheres	Oak Ridge National Laboratory
Glass Marbles in a Lead Matrix	Pacific Northwest Laboratory

Screening Process

The first input considered in the evaluation of potential waste forms for immobilization of SRP high-level waste was a series of preliminary product and process evaluations[4-7] conducted by each of the DOE defense sites (Savannah River, Hanford, and Idaho) to determine the preferred forms for immobilization of the high-level waste existing at each specific site. Additionally, two studies[8,9] were conducted by Pacific Northwest Laboratory (PNL) to assess potential commercial waste forms and processes. Borosilicate glass was consistently the highest ranked form in each evaluation for immobilizing both defense and commercial high-level waste.* Either ceramic forms or other glass forms were the second most-preferred forms.

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* In this discussion, rank and rate have the following meanings: rank is used in the sense of an ordinal number giving relative standing or position;

rate is used to refer to a numerical value obtained through an evaluation or grading process.

As a second input, an Alternative Waste Form Peer Review Panel has been convened annually[1,2] since 1979 to review the relative scientific merits and engineering practicality of high-level waste forms being developed. The panel's most recent review[10] in May 1981 produced a relative ranking of the seven candidate forms. Borosilicate glass was ranked as the preferred form for immobilization of high-level waste followed in order by Synroc, high-silica glass, tailored ceramic, coated particles, FUETAP concrete, and glass marbles in a lead matrix.

A quantitative evaluation of waste form performance, the third input, was performed by the Savannah River Laboratory (SRL) using a rating system similar to one developed by a DOE Interface Working Group on High-Level Waste Form Selection Factors.[11] The evaluation compared the seven candidate waste forms on the basis of waste loading, mechanical stability, and leach resistance, with leach resistance given the highest weight. Waste loading was defined as curie content of SRP high-level waste per unit volume of waste form; mechanical stability was inferred from standard impact tests at Argonne National Laboratory; and leach resistance was determined by use of standard leach testing procedures developed by the Materials Characterization Center (MCC). Leaching data were provided by the waste form developers, MCC, and SRL.

Based on this evaluation,[3] the waste forms were divided into three groups: (1) Synroc, tailored ceramic, and coated particles had the highest ratings; (2) borosilicate glass and high-silica glass had intermediate ratings; and (3) glass marbles in a lead matrix and FUETAP concrete had the lowest ratings. A clear delineation based on product performance could be made between the highest and lowest rated waste forms; distinctions between waste forms in the high and intermediate categories were less clear. The ceramic forms rated highest because they had the lowest uranium leach rates (the highest weighted single property); however, the glass forms rated better than the ceramics when considering leach rates for cesium and strontium (the main contributors to the curie content of the waste). Delineation among waste forms within a particular group was not possible based on product performance alone.

The fourth input was a processability analysis conducted by the Engineering Department of E. I. du Pont de Nemours and Company.[12] To evaluate quantitatively the waste form processes, twenty-one processability criteria were developed in four major categories: reliability/complexity, resource requirements, personnel safety, and quality control. Process data evaluated against these criteria were obtained from process flowsheets, equipment definitions, and conceptual facility layouts developed in collaboration with SRL and each of the waste form developers. The ratings based on processability fell in four general groups:

borosilicate glass and FUETAP concrete, relatively simple; glass marbles in a lead matrix and high-silica glass, moderately complex; crystalline ceramics, complex; and coated sol-gel particles, very complex.

Waste form ratings from the product performance and processability evaluations were combined to obtain an overall ranking of the seven waste forms. The ranking in order of highest to lowest was: borosilicate glass, Synroc and tailored ceramic, high-silica glass, FUETAP concrete, coated particles, and glass marbles in a lead matrix. Generally waste forms with high product performance ratings had low processability ratings, and vice versa. Borosilicate glass achieved the highest overall ranking because it had the highest processability rating combined with an intermediate product rating. The two ceramic forms ranked second overall because their high product ratings compensated for their lower processability ratings.

Screening Results

Based on the results of each of the four major inputs discussed above, borosilicate glass and crystalline ceramic were selected in November 1981 for further development as potential waste forms for immobilization of SRP high-level waste.

Borosilicate glass was selected for continued development on the following bases:

- Borosilicate glass demonstrated acceptable product performance properties.
- Borosilicate glass was ranked as the preferred form for high-level waste immobilization by the Alternative Waste Form Peer Review Panel.
- Borosilicate glass was consistently selected as the preferred form by the DOE defense sites, and was rated highest in the commercial waste form evaluations.
- The process for fabricating the borosilicate glass waste form is the simplest and least expensive of all those considered.

The crystalline ceramic forms, although ranking rather low in processing, were selected as the best alternative to borosilicate glass on the following bases:

- The crystalline ceramic forms, Synroc and tailored ceramic, ranked highest in the product performance evaluation.

- The Synroc form, ranked second by the Alternative Waste Form Peer Review Panel, was judged to be the best characterized and understood of the forms other than borosilicate glass.
- Ceramic waste forms consistently ranked high in each of the DOE defense-site evaluations.
- The ceramics have generally better high-temperature leaching characteristics than borosilicate glass.
- A number of mineral analogues of the crystalline ceramics have proven extremely durable in nature.

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APPENDIX C

DESCRIPTION OF SRP HIGH-LEVEL WASTE

DESCRIPTION OF SRP HIGH-LEVEL WASTE

Description

Chemical separations of irradiated fuel and targets at SRP result in product streams and acidic waste streams that contain almost all of the fission products, and small amounts of unrecovered uranium and transuranics. This acidic waste stream is made alkaline (pH 10 to 13) by addition of sodium hydroxide and transferred to large (about 4,900 m^3) underground storage tanks with multiple barriers of carbon steel and reinforced concrete.

In the waste storage tanks, components insoluble in the highly alkaline solution precipitate and settle to form a layer of flocculent sludge on the tank bottom. Most of the radioactive elements, including strontium and the actinides, are contained in the sludge; only the cesium remains predominantly in the supernatant liquid. Settled sludge volume is from 4 to 7% of the waste received, but 70 to 90% of this volume is interstitial liquid with a composition similar to the supernate.

After one to two years' storage, radioactivity of short-lived fission products has largely decayed, and the diminished thermal agitation permits most of the suspended sludge and associated radioactive components to settle out. Then the supernatant liquid, containing most of the soluble, nonradioactive salts and the radioactive cesium, is decanted off to other waste tanks and processed through evaporators to remove most of the water.

The partially dewatered waste concentrate from the evaporators is discharged to waste tanks while hot. On cooling, part of the dissolved salt mixture (chiefly sodium nitrate, nitrite, carbonate, sulfate, and hydroxide) crystallizes out of solution and deposits in the tank as damp salt cake. The remaining supernatant liquid is recycled back to the evaporators for removal of more water and additional crystallization of salt cake.

About 110,000 m^3 (28 million gallons) of high-level waste are presently stored at SRP. The actual volumes at any time in the future will be a function of the waste generation from plant operations, DWPF startup, and the operations to concentrate the waste.

The sludge (containing most of the strontium-90 and the actinides) will be the initial feed to the DWPF. High-activity components from the supernate (primarily cesium-137 and small amounts of strontium and the actinides) will be concentrated in another facility for mixing with the sludge feed to the DWPF or recovered for beneficial use. Continued development of supernate

processing technology is expected to reduce significantly the cost and complexity of the supernate decontamination and disposal process described in the DWPF EIS.[1]

The sludge characteristics will determine the composition and properties of the waste form. The composition of the existing sludge varies considerably from tank to tank and, to a lesser extent, within each tank. Principal elements of the sludge measured from samples taken from several tanks are listed in Table C-1. The effects of waste composition on glass product performance have been studied with simulated waste glass,[2] and acceptable performance has been obtained with compositional variations more extreme than expected in practice.

Heat Generation

SRP waste storage tanks now contain about 2.6 MW of heat-generating fission products. The major contributors are:

Isotope	Megawatts	Half-Life, yr
Cs-137	0.63	30.0
Sr-90	0.77	28.0
Ce-144	0.63	0.78
Misc.	0.53	-

Without replenishment from fresh waste, heat generation from Ce-144 in the stored waste will disappear within 3 to 4 years, and the miscellaneous contributions from short-lived fission products, such as Cs-134, Ru-106, and Pm-147, will decay away within 10 years. Therefore, decay of Sr-90 and Cs-137 would be the major source of heat generation from DWPF waste canisters in a geologic repository.

Currently the contribution of Cs-137 and Sr-90 in fresh waste generated annually corresponds to about 4% of the existing inventory in SRP waste tanks; however, about 2% of this inventory decays each year. Consequently, the heat generation rate of fission products requiring geologic disposal is increasing about 2% annually. Based on current projections of future operations, heat generation from Cs-137 and Sr-90 in stored SRP high-level waste is not expected to exceed 2.0 MW (by the year 2000, the rate of accumulation is expected to be equaled by the rate of decay).

TABLE C-1

Compositional Variations in SRP Waste Sludge

Element*	Amount, wt % Tanks 4 and 6	Tank 5	Tank 13	Tank 15	Tank 16
Fe	32.8	28.9	25.6	5.3	13.9
Al	2.3	1.6	8.7	18.8	16.6
Mn	2.0	5.8	7.8	2.4	2.6
U	9.2	10.8	4.2	3.8	4.5
Na	3.0	5.7	2.6	2.4	2.2
Ca	2.3	0.9	1.8	0.5	2.9
Ni	6.3	6.3	0.4	0.7	0.3

* Present as components with other elements such as oxygen, hydrogen, nitrogen, and sulfur.

Although Pu-238 contributes only 0.5% of the curie content of SRP high-level waste, it will contribute about 8% to the heat generation in canisters. Assuming that the current 8% contribution will continue, the total heat generation in DWPF waste canisters in a geologic repository (containing SRP high-level waste existing and produced over the next two decades) is estimated to be 2.2 MW.

Consequently, if borosilicate glass is selected as the waste form, the average heat generation rate of a DWPF borosilicate glass canister would be about 220 watts based on the production of 10,000 waste canisters. Because of tank-to-tank variations in waste composition, and because of changes in the reference process that may result from ongoing development, the maximum heat generation rate in the DWPF canisters will vary. However, the production techniques can be utilized to limit canister heat generation rates to level within applicable regulatory requirements.

For design purposes (e.g., establishing shielding requirements in the DWPF), the reference DWPF borosilicate glass waste canister is assumed to contain 150,000 Ci of radionuclides and to generate 423 watts.[1] Based upon the projected maximum of 2.2 MW in SRP high-level waste canisters, the average heat generation of about 220 watts per DWPF waste canister will be well below the design basis value and even further below the typical heat rating of canisters containing commercial high-level waste (Table 2.1 of Reference 1).

Calculated surface temperatures of the reference DWPF borosilicate glass waste canister in a salt repository are shown in Figure C-1. The maximum surface temperature occurs approximately 20 years after the waste is emplaced and will be about 80°C in salt[2] and somewhat higher in rock repositories such as granite and basalt. The calculations assume that the canister is generating 256 watts when emplaced in the repository (i.e., 10 years after the reference canister is produced). After the 1000-year containment period (Section 3.2.5.3), waste form surface temperatures would be at ambient repository temperatures; e.g., about 20°C for granite, 35°C for salt, and about 50°C for basalt.

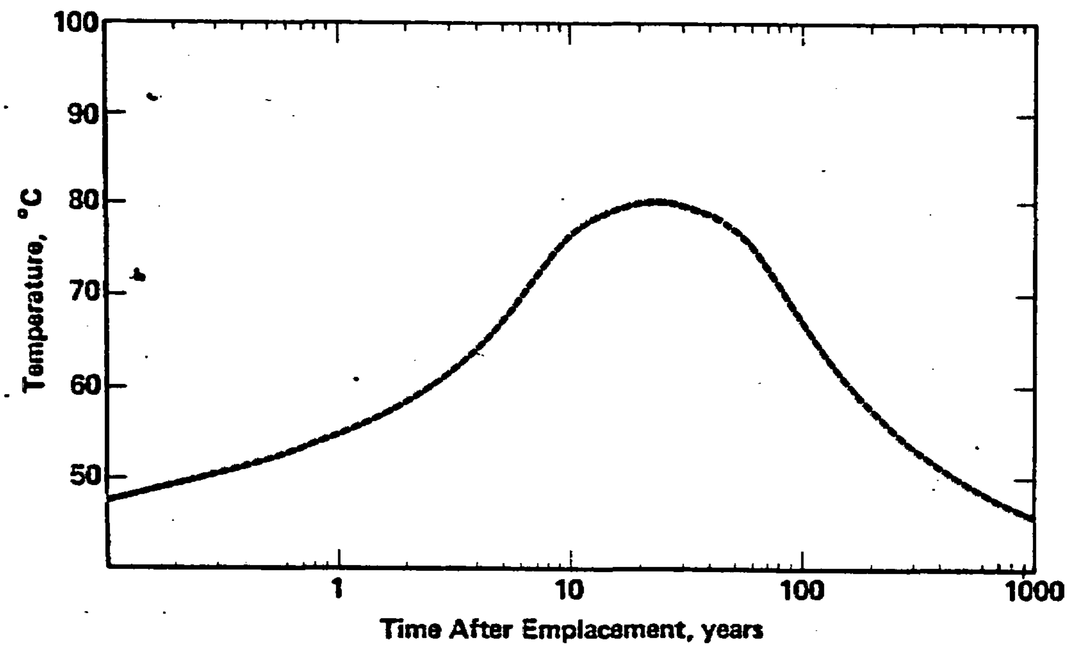

FIGURE C-1. Surface Temperatures for a Reference DWPF Borosilicate Glass Canister in a Salt Repository

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APPENDIX D

GENERIC REPOSITORY DESCRIPTIONS

Candidate Geologic Media

The rock types being studied as potential repository media include salt, basalt, and tuff. Crystalline rocks have also reeived some attention. Bedded salt is found in Utah (Paradox Basin); New Mexico and Texas (Permian Basin); and in Michigan, Ohio, Pennsylvania, Kansas, Oklahoma, and New York (Salina Basin). Salt domes are located in Mississippi, Louisiana, and Texas. Basalt has been studied at the DOE Hanford Reservation in Washington, and tuff at the Nevada Test Site (Figure D-1).

Each type of rock has properties that are considered important for waste containment. Salt is being studied because of its long-term stability, strength, and heat-dissipating characteristics. Basalt appears suitable because of its strength, thermal conductivity, and expansion properties. Tuff is being studied because of its strength, high sorptive qualities, and location adjacent to other sorptive strata.

Bedded Salt. Bedded salt[1] occurs in multiple horizontal strata, separated by strata of other minerals. A single salt bed may be as much as 60 meters thick and many kilometers wide in the horizontal directions. The thick salt beds are surrounded by thinner, more porous and permeable strata. Overlying much of the salt-bearing section are sediments which locally serve as aquifers.

Salt beds considered to be of possible interest as repository sites are required to be at least 21 m thick, to contain at least 85 per cent salt, to have no non-salt interbeds thicker than 3 m, and to lie between 300 and 900 m below the surface.

The presence of aquifers above the repository level requires special seals along the vertical shafts and horizontal tunnels.[2] Seals include relatively impermeable bulkheads keyed into the walls to intercept flow of water, and backfills such as concretes, clays, and crushed salt.

Domed Salt. Salt domes are very large vertically oriented extrusions of underlying deposits of essentially pure halite, principally occurring in the Gulf Coast region.[3] No two domes are alike. The horizontal cross section of a typical dome is slightly elliptical, having dimensions of 3000 m by 4000 m and having its greatest diameters at depths of 600 to 1200 m below the surface.

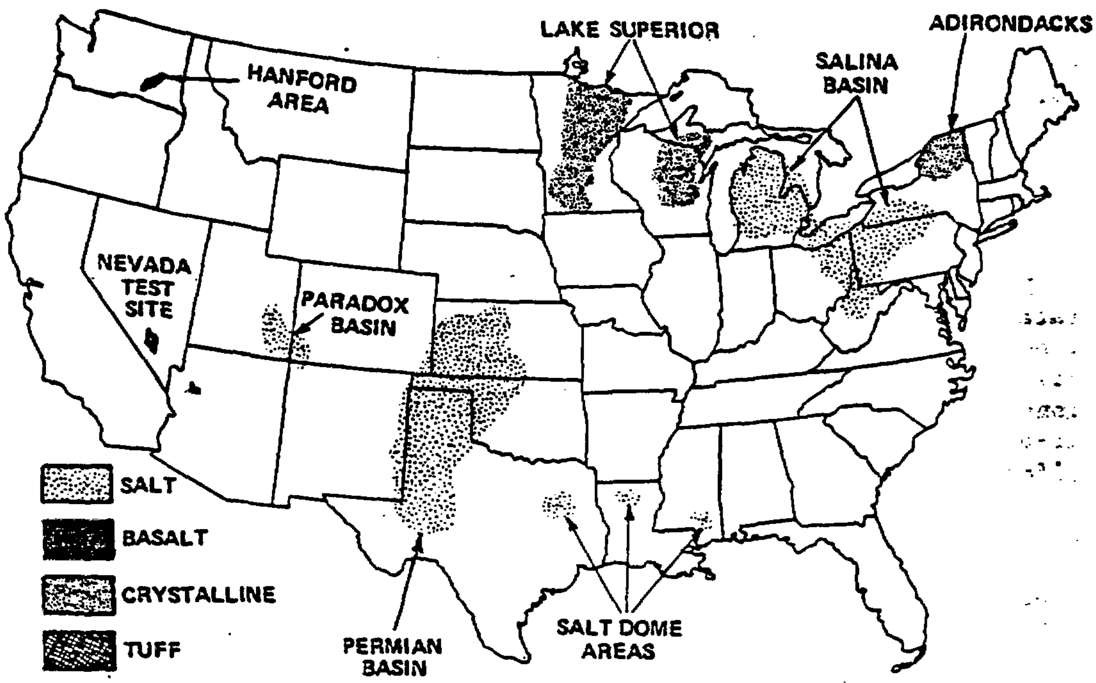

FIGURE D-1. Possible Locations of Geologic Repositories in Various Types of Rock

The top of the salt dome is typically about 400 m below the land surface and is overlain with about 30 m of caprock consisting of limestone and anhydrite. Strata along the flanks are swept up near the dome, and may be faulted with as much as 60 m of offset. Aquifers overlie and are adjacent to the flanks of the dome. The typical dome is overlain by several sedimentary formations.

Basalt. The Columbia Plateau basalts are igneous rock composed of individual lava flows, layered one on top of the other, which extend to depths of approximately 2700 m beneath the land surface at the Hanford Reservation, near Richland, WA.

The one site under consideration for a possible repository is the Umtanum Flow, 15 million years old, 45 to 60 m thick, and 1100 m below the surface.[4] The interior of the Umtanum has a glass-rich texture and secondary mineral content which provides sorptive minerals and alteration products along potential groundwater pathways. The hydrology of this area has been studied extensively down to 1370 m below ground level.

Tuff. Detailed investigations are in progress to determine the suitability of tuffs at Yucca Mountain, in the southwest corner of the U.S. DOE Nevada Test Site, for storage of high-level radioactive waste.[5] A site selection committee is scheduled to provide a specific recommendation by December 1982.

Repository Arrangement

The three main components of the generic repository, the underground facilities, the shafts, and the surface facilities are illustrated in the cutaway view shown in Figure D-2. The underground surface area is 840 hectares (ha), including the buffer zone, and the net underground working area is approximately 560 ha.

The shafts are drilled in a protective zone of the host rock about 640 meters on a side, called the shaft pillar. Access to the repository is through 5 shafts bored in the shaft pillar, which[3] is located centrally to equalize any thermal loads on the shafts and to minimize haul distances for both the excavated rock and the waste. The shaft pillar provides structural support for the shafts and contains facilities required for underground development and waste emplacement operations. All shafts are lined with steel and grouted to about 30 m into host rock formation.

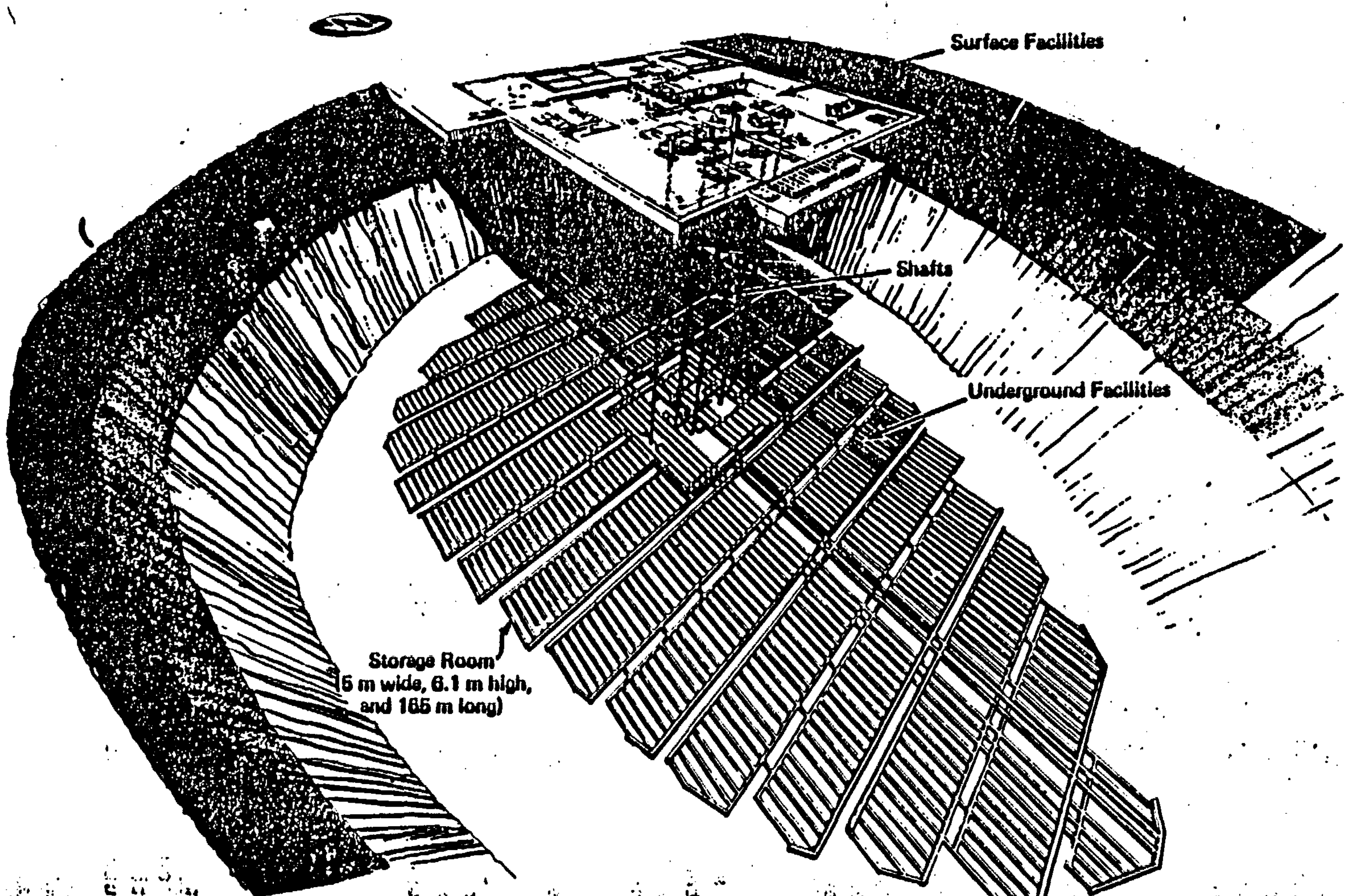

FIGURE D-2. Generic Repository[3]

Corridors, 9 m wide and 5 m high, extending from the principal axis of the underground facility provide for movement of mining equipment, rock conveyors, waste and materials movement, and the ventilation systems. Branch corridors at right angles to the main corridors provide access to the waste storage rooms. Storage rooms are 5 m wide by 6 m high by 165 m long, separated by 21-m-wide support pillars. The waste canisters are emplaced in pre-drilled holes in the floor of the storage room. The spacing of holes depends on mechanical limitations and the thermal characteristics of the waste.

The most important surface facilities are located in the exclusion area, directly over the shaft pillar. In the waste-handling building, waste canisters are received, unloaded from shipping casks, overpacked if necessary, and transferred to the terminal storage area. This building is built over the waste transfer shaft. Auxiliary facilities include four additional shafts which combine functions for men and materials handling, rock handling, and underground ventilation.

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GLOSSARY OF ACRONYMS

ANL - Argonne National Laboratory

ATLAS - A half-scale prototype AVM

AVH - Ateliers de vitrification de La Hague (vitrification facilities at La Hague)

AVM - Ateliers de vitrification de Marcoule (vitrification facilities at Marcoule)

BNFL - British Nuclear Fuels, Ltd.

CFR - Code of Federal Regulations

COGEMA - Compagnie Generale des Matieres Nucleaires (France's commercial fuel reprocessing company)

CRRD - Conceptual Reference Repository Description

DOE - Department of Energy

DOT - Department of Transportation

DWK - Deutsche Gesellschaft für Wideraufbeitung von Kernbrennstoffen mbH (German fuel reprocessing company)

DWPF - Defense Waste Processing Facility

EIS - Environmental Impact Statement

EPA - Environmental Protection Agency

FINGAL - Early U.K. rising level, pot-vitrification process

FUETAP - Formed under elevated temperature and pressure

HARVEST - Recent U.K. rising level, pot-vitrification process

HEPA - High efficiency particulate air

HIP - Hot isostatic press

HOVA - Vitrification plant at WAK pilot plant

LLNL - Lawrence Livermore National Laboratory

GLOSSARY OF ACRONYMS, Contd

MCC	- Materials Characterization Center
MTHM	- Metric tons of heavy metal
NEPA	- National Environmental Policy Act
NRC	- Nuclear Regulatory Commission
NWTS	- National Waste Terminal Storage
PAMELA	- Belgium/German Vitrification process to produce glass blocks or beads
PIVER	- French hot vitrification pilot plant
PNL	- Pacific Northwest Laboratory
SRL	- Savannah River Laboratory
SRP	- Savannah River Plant
WAK	- Wiederaufarbeitungsanlage Karlsruhe Betriebgesselsdraft mbH (German fuel reprocessing company at Karlsruhe)