

IV. BACKGROUND AND DESCRIPTION OF ALTERNATIVES

A. DESCRIPTION OF SRP HIGH-LEVEL LIQUID WASTE

1. Characteristics of Waste

Almost all (>99%) of the fission products generated in the fuel during reactor operations go into acidic aqueous waste streams during spent fuel processing. These wastes are made alkaline to a pH of 10 to 13 and transferred to large underground waste storage tanks. In the waste storage tanks, components insoluble in the highly alkaline solution precipitate and settle to form a layer of sludge on the tank bottom. The sludge contains oxides and/or hydroxides of manganese, iron, silicon, and aluminum, along with fission products, induced radioactive elements, uranium, transuranium elements, mercury, silver, and other nonradioactive elements. Most of the radionuclides are contained in the sludge; only the cesium remains predominantly in the liquid. Settled sludge volume is 6 to 10% of the total (unevaporated) waste volume, but 70 to 90% of this volume is interstitial liquid with a composition similar to the supernatant liquid. After the sludge settles to the bottom of a tank, the supernatant liquid is transferred to an evaporator for dewatering. The concentrate from the evaporator is transferred to a cooled waste tank where the supersaturated solution precipitates and forms salt crystals. The supernate is returned to the evaporator for further concentration. This process is repeated until essentially all of the supernate is converted to damp salt cake.

The waste in a single tank is made up of many waste streams from the spent fuel reprocessing plants, and its detailed composition varies from tank to tank. The chemical composition of the major components of the composite fresh waste is given in Table IV-1. Table IV-2 shows the concentrations of radionuclides in the fresh waste, with the assumption that the fuel has been cooled six months before being reprocessed. The radionuclide concentration in the salt is approximately three times the concentration in supernate with the same decay period.

TABLE IV-1

Average Chemical Composition of Fresh
SRP High-Level Waste

| <i>Constituent</i> | <i>Concentration</i> | |
|---------------------------------|----------------------|------------|
| | <i>Molar</i> | <i>g/L</i> |
| NaNO ₃ | 3.3 | 281 |
| NaNO ₂ | <0.2 | <14 |
| NaAl(OH) ₄ | 0.5 | 59 |
| NaOH | 1 | 40 |
| Na ₂ CO ₃ | 0.1 | 11 |
| Na ₂ SO ₄ | 0.3 | 43 |
| Fe(OH) ₃ | 0.07 | 7.5 |
| MnO ₂ | 0.02 | 1.7 |
| Hg(OH) ₂ | 0.002 | 0.5 |
| Other Solids | 0.13 ^a | 7.8 |

a. Assuming an average molecular weight
of 60.

TABLE IV-2

Average Radionuclide Composition of Fresh^a SRP High-Level Waste

| <i>Radionuclide</i> | <i>Activity, Ci/gal</i> | <i>Radionuclide</i> | <i>Activity, Ci/gal</i> |
|--------------------------------------|-------------------------|--------------------------------------|-------------------------|
| ⁹⁵ Nb | 105 | ²⁴¹ Am | 1 × 10 ⁻³ |
| ¹⁴⁴ Ce- ¹⁴⁴ Pr | 68 | ⁹⁹ Tc | 5 × 10 ⁻⁴ |
| ⁹⁵ Zr | 60 | ²³⁹ Pu | 3 × 10 ⁻⁴ |
| ⁹¹ Y | 47 | ¹⁵⁴ Eu | 1 × 10 ⁻⁴ |
| ⁸⁹ Sr | 36 | ⁹³ Zr | 1 × 10 ⁻⁴ |
| ¹⁴¹ Ce | 12 | ²⁴⁰ Pu | 6 × 10 ⁻⁵ |
| ¹⁴⁷ Pm | 12 | ¹³⁵ Cs | 4 × 10 ⁻⁵ |
| ¹⁰³ Ru | 10 | ¹²⁶ Sn- ¹²⁶ Sb | 1 × 10 ⁻⁵ |
| ¹⁰⁶ Ru- ¹⁰⁶ Rh | 4 | ⁷⁹ Se | 1 × 10 ⁻⁵ |
| ⁹⁰ Sr | 3 | ²³³ U | 2 × 10 ⁻⁶ |
| ¹³⁷ Cs | 3 | ¹²⁹ I | 1 × 10 ⁻⁶ |
| ¹²⁹ Te | 2 | ²³⁸ U | 6 × 10 ⁻⁷ |
| ¹²⁷ Te | 2 | ¹⁰⁷ Pd | 5 × 10 ⁻⁷ |
| ¹³⁴ Cs | 1 | ²³⁷ Np | 4 × 10 ⁻⁷ |
| ¹⁵¹ Sm | 8 × 10 ⁻² | ¹⁵² Eu | 2 × 10 ⁻⁷ |
| ²³⁸ Pu | 1 × 10 ⁻² | ²⁴² Pu | 6 × 10 ⁻⁸ |
| ²⁴¹ Pu | 2 × 10 ⁻³ | ¹⁵⁸ Tb | 6 × 10 ⁻⁸ |
| ²⁴⁴ Cm | 1 × 10 ⁻³ | ²³⁵ U | 3 × 10 ⁻⁸ |

a. After reprocessing fuel that has been cooled six months after discharge from reactor. See Table IV-6 for the average radionuclide concentration of reconstituted SRP high-level waste in 1985.

Both the chemical and radionuclide composition of the waste changes as the waste ages. The major changes are:

- Radiolytic decomposition of the waste. The major effect of this radiolytic decomposition is the slow reduction in the NaNO_3 concentration with an equivalent increase in NaNO_2 concentration. After 5 to 10 years, the NaNO_2 concentration approaches the residual NaNO_3 concentration.
- A slow reduction in the NaOH concentration due to reaction with CO_2 absorbed from air, forming Na_2CO_3 .
- Decay of radionuclides. Figure IV-1 shows the decay of major radionuclides in this waste.
- Natural partitioning of the waste into sludge and supernate fractions. The sludge scavenges most of the radionuclides from the supernate as it settles to the bottom of the tank.

2. Characteristics of Reconstituted Waste for Long-Term Management

If waste removal from tanks is initiated in 1988, 25 waste tanks are expected to be in service. These tanks will contain approximately 12.7 million gallons (47 million liters) of damp crystallized salt, 3 million gallons (15 million liters) of sludge, and 6.2 million gallons (22 million liters) of liquid waste. Tables IV-3 and IV-4 show their radionuclide compositions with age.

Before solidification of the waste is started (or before transfer to an SRP bedrock cavern or replacement of waste storage tanks), the salt must be dissolved and the solution is used to slurry the sludge from the waste tanks. Dissolution of the total salt expected to be on hand in 1987 will require approximately 40 million gallons (150 million liters) of water. These operations will produce about 60 million gallons (227 million liters) of reconstituted waste which will be fed to the solidification facility or SRP bedrock cavern or returned to new waste storage tanks.

The reconstituted waste will be similar in chemical composition to the original neutralized fresh waste generated by the spent fuel reprocessing plants but will be less radioactive. The chemical composition of the reconstituted waste is shown in Table IV-5. Table IV-6 gives the activity of the significant radionuclides in reconstituted waste. Figures IV-2 and IV-3 show the radionuclide content of the waste from 0 to 1400 years and from 0 to 10^6 years after irradiation, respectively. The units are expressed as Ci/gal of reconstituted waste.

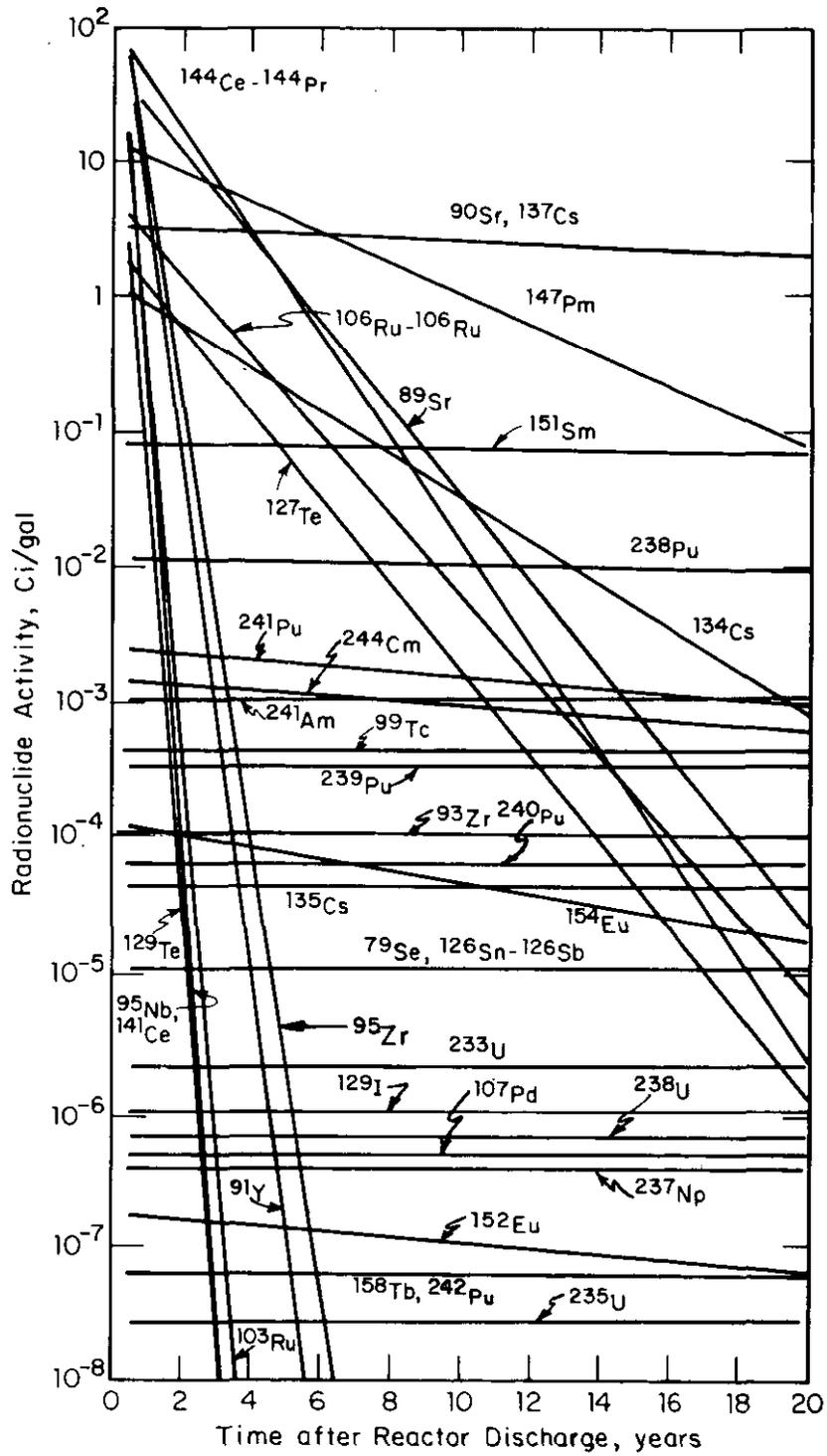


FIGURE IV-1. Radionuclide Composition of SRP Waste (0 to 20 years after irradiation)

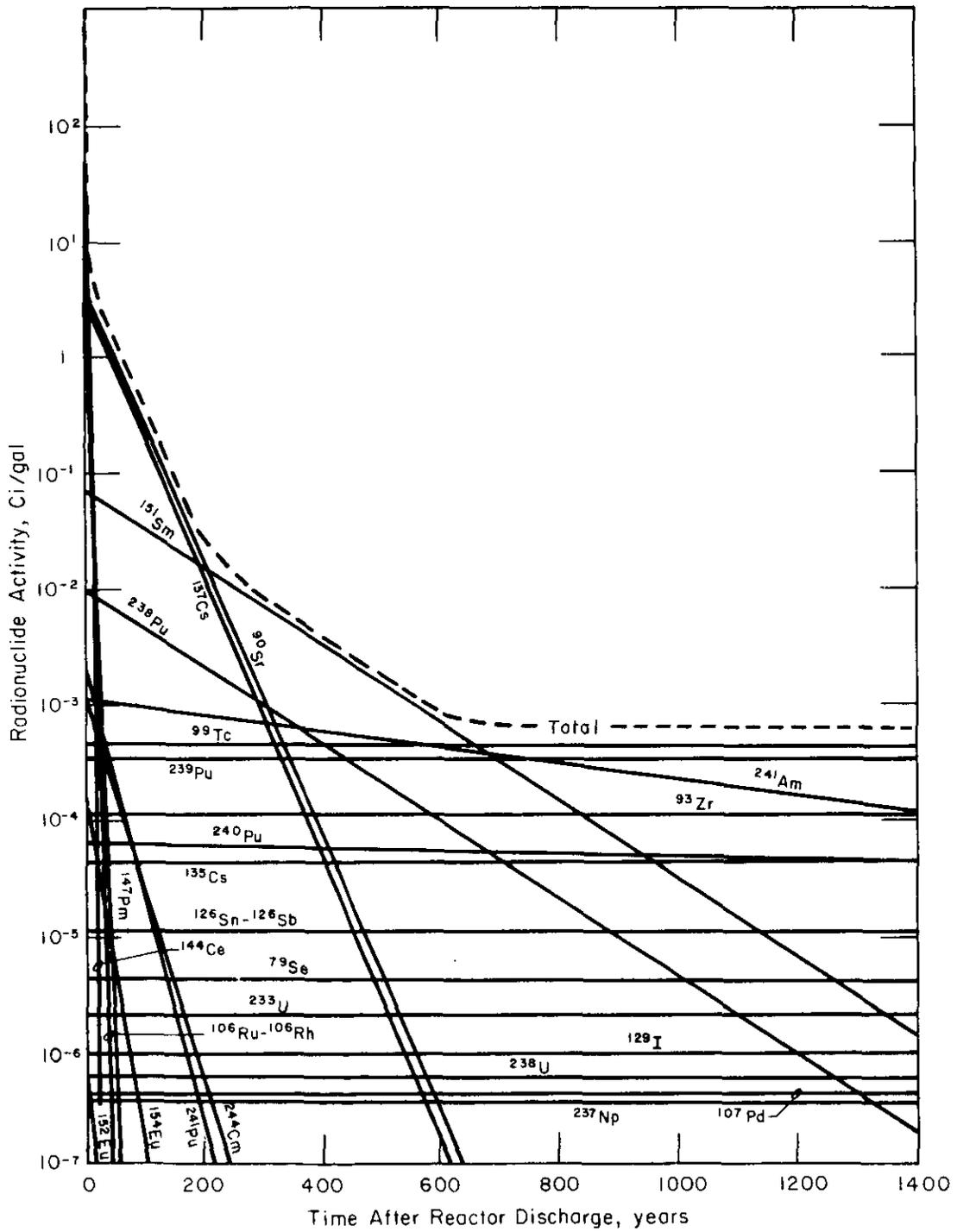


FIGURE IV-2. Radionuclide Composition of SRP Waste (0 to 1400 years after irradiation)

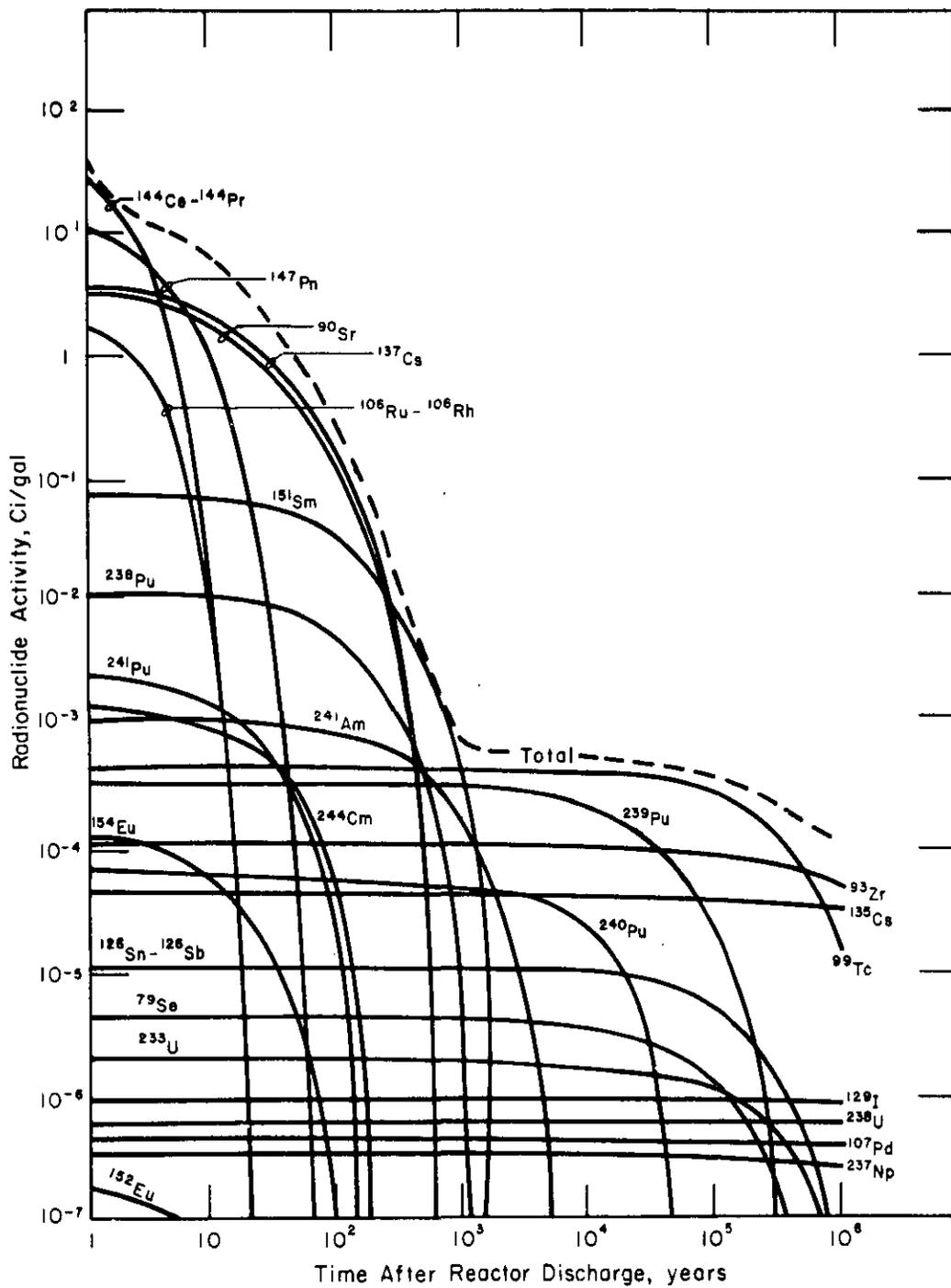


FIGURE IV-3. Radionuclide Composition of SRP Waste (1 to 10⁶ years after irradiation)

TABLE IV-3

Average Radionuclide Composition of SRP High-Level Sludge

| Time After Irradiation, years → | Radionuclide Activity, Ci/gal | | | | Radionuclide Activity, Ci/gal | | |
|--------------------------------------|-------------------------------|------------------------|------------------------|--------------------------------------|-------------------------------|------------------------|------------------------|
| | 1 | 5 | 10 | | 1 | 5 | 10 |
| ¹⁴⁴ Ce- ¹⁴⁴ Pr | 4.5 x 10 ² | 1.3 x 10 ¹ | 1.5 x 10 ⁻¹ | ²⁴¹ Am | 1.1 x 10 ⁻² | 1.1 x 10 ⁻² | 1.1 x 10 ⁻² |
| ⁹⁵ Zr | 9.6 x 10 ¹ | 1.8 x 10 ⁻⁵ | a | ⁹⁹ Tc | 4.3 x 10 ⁻³ | 4.3 x 10 ⁻³ | 4.3 x 10 ⁻³ |
| ⁹⁴ Y | 7.6 x 10 ¹ | 2.5 x 10 ⁻⁶ | a | ²³⁹ Pu | 3.5 x 10 ⁻³ | 3.5 x 10 ⁻³ | 3.5 x 10 ⁻³ |
| ⁸⁹ Sr | 4.4 x 10 ¹ | a | a | ¹⁵⁴ Eu | 1.1 x 10 ⁻³ | 8.3 x 10 ⁻⁴ | 5.5 x 10 ⁻⁴ |
| ⁹⁵ Nb | 6.0 x 10 ⁰ | a | a | ⁹³ Zr | 8.6 x 10 ⁻⁴ | 8.6 x 10 ⁻⁴ | 8.6 x 10 ⁻⁴ |
| ¹⁴¹ Ce | 3.0 x 10 ⁰ | a | a | ²⁴⁰ Pu | 6.4 x 10 ⁻⁴ | 6.4 x 10 ⁻⁴ | 6.4 x 10 ⁻⁴ |
| ¹⁴⁷ Pm | 1.0 x 10 ² | 3.6 x 10 ¹ | 9.7 x 10 ⁰ | ¹³⁵ Cs | 2.2 x 10 ⁻⁵ | 2.2 x 10 ⁻⁵ | 2.2 x 10 ⁻⁵ |
| ¹⁰³ Ru | 5.2 x 10 ⁰ | a | a | ¹²⁶ Sn- ¹²⁶ Sb | 1.1 x 10 ⁻⁴ | 1.1 x 10 ⁻⁴ | 1.1 x 10 ⁻⁴ |
| ¹⁰⁶ Ru- ¹⁰⁶ Rh | 2.4 x 10 ¹ | 1.6 x 10 ⁰ | 5 x 10 ⁻² | ⁷⁹ Se | 1.0 x 10 ⁻⁴ | 1.0 x 10 ⁻⁴ | 1.0 x 10 ⁻⁴ |
| ⁹⁰ Sr | 3.0 x 10 ¹ | 2.8 x 10 ¹ | 2.4 x 10 ¹ | ²³³ U | 2.1 x 10 ⁻⁵ | 2.1 x 10 ⁻⁵ | 2.1 x 10 ⁻⁵ |
| ¹³⁷ Cs | 1.6 x 10 ⁰ | 1.5 x 10 ⁰ | 1.3 x 10 ⁰ | ¹²⁹ I | 9.4 x 10 ⁻⁶ | 9.4 x 10 ⁻⁶ | 9.4 x 10 ⁻⁶ |
| ¹²⁹ Te | 9.4 x 10 ⁻¹ | a | a | ²³⁸ U | 6.4 x 10 ⁻⁶ | 6.4 x 10 ⁻⁶ | 6.4 x 10 ⁻⁶ |
| ¹²⁷ Te | 6.4 x 10 ⁰ | 5.9 x 10 ⁻⁴ | a | ¹⁰⁷ Pd | 4.4 x 10 ⁻⁶ | 4.4 x 10 ⁻⁶ | 4.4 x 10 ⁻⁶ |
| ¹³⁴ Cs | 8.7 x 10 ⁰ | 2.3 x 10 ⁰ | 4.2 x 10 ⁻¹ | ²³⁷ Np | 3.9 x 10 ⁻⁶ | 3.9 x 10 ⁻⁶ | 3.9 x 10 ⁻⁶ |
| ¹⁵¹ Sm | 7.5 x 10 ⁻¹ | 7.3 x 10 ⁻¹ | 7.0 x 10 ⁻¹ | ¹⁵² Eu | 1.7 x 10 ⁻⁶ | 1.3 x 10 ⁻⁶ | 1.0 x 10 ⁻⁶ |
| ²³⁸ Pu | 1.1 x 10 ⁻¹ | 1.1 x 10 ⁻¹ | 1.1 x 10 ⁻¹ | ²⁴² Pu | 6.2 x 10 ⁻⁷ | 6.2 x 10 ⁻⁷ | 6.2 x 10 ⁻⁷ |
| ²⁴¹ Pu | 2.4 x 10 ⁻² | 2.0 x 10 ⁻² | 1.6 x 10 ⁻² | ¹⁵⁸ Tb | 6.0 x 10 ⁻⁷ | 6.0 x 10 ⁻⁷ | 6.0 x 10 ⁻⁷ |
| ²⁴⁴ Cm | 1.3 x 10 ⁻² | 1.1 x 10 ⁻² | 9.5 x 10 ⁻² | ²³⁵ U | 2.7 x 10 ⁻⁷ | 2.7 x 10 ⁻⁷ | 2.7 x 10 ⁻⁷ |

a. Value <1 x 10⁻⁷.

TABLE IV-4

Average Radionuclide Composition of SRP High-Level Supernate

| Time After Irradiation, years → | Radionuclide Activity, Ci/gal | | | | Radionuclide Activity, Ci/gal | | |
|--------------------------------------|-------------------------------|------------------------|------------------------|--------------------------------------|-------------------------------|------------------------|------------------------|
| | 1 | 5 | 10 | | 1 | 5 | 10 |
| ¹⁴⁴ Ce- ¹⁴⁴ Pr | 2.6 | 7.4 x 10 ⁻² | 8.7 x 10 ⁻⁴ | ²⁴¹ Am | 3.6 x 10 ⁻⁶ | 3.6 x 10 ⁻⁶ | 3.6 x 10 ⁻⁶ |
| ⁹⁵ Zr | 2.7 | 5.0 x 10 ⁻⁷ | a | ⁹⁹ Tc | 2.5 x 10 ⁻⁵ | 2.5 x 10 ⁻⁵ | 2.5 x 10 ⁻⁵ |
| ⁹¹ Y | 1.7 x 10 ⁻¹ | 5.7 x 10 ⁻⁹ | a | ²³⁹ Pu | 1.1 x 10 ⁻⁶ | 1.1 x 10 ⁻⁶ | 1.1 x 10 ⁻⁶ |
| ⁸⁹ Sr | 1.0 x 10 ⁻¹ | a | a | ¹⁵⁴ Eu | 6.7 x 10 ⁻⁶ | 4.8 x 10 ⁻⁶ | 3.2 x 10 ⁻⁶ |
| ⁹⁸ Nb | 1.7 x 10 ⁻¹ | a | a | ⁹³ Zr | 2.4 x 10 ⁻⁵ | 2.4 x 10 ⁻⁵ | 2.4 x 10 ⁻⁵ |
| ¹⁴¹ Ce | 1.7 x 10 ⁻¹ | a | a | ²⁴⁰ Pu | 2.1 x 10 ⁻⁷ | 2.1 x 10 ⁻⁷ | 2.1 x 10 ⁻⁷ |
| ¹⁴⁷ Pm | 6.1 x 10 ⁻¹ | 2.1 x 10 ⁻¹ | 5.7 x 10 ⁻² | ¹³⁵ Cs | 4.6 x 10 ⁻⁵ | 4.6 x 10 ⁻⁵ | 4.6 x 10 ⁻⁵ |
| ¹⁰³ Ru | 1.4 x 10 ⁻¹ | a | a | ¹²⁶ Sn- ¹²⁶ Sb | 6.1 x 10 ⁻⁷ | 6.1 x 10 ⁻⁷ | 6.1 x 10 ⁻⁷ |
| ¹⁰⁶ Ru- ¹⁰⁶ Rh | 6.7 x 10 ⁻¹ | 4.3 x 10 ⁻² | 1.4 x 10 ⁻³ | ⁷⁹ Se | 6.0 x 10 ⁻⁷ | 6.0 x 10 ⁻⁵ | 6.0 x 10 ⁻⁷ |
| ⁹⁰ Sr | 6.8 x 10 ⁻² | 6.2 x 10 ⁻² | 5.5 x 10 ⁻² | ²³³ U | 7.1 x 10 ⁻⁹ | 7.1 x 10 ⁻⁹ | 7.1 x 10 ⁻⁹ |
| ¹³⁷ Cs | 3.3 | 3.1 | 2.7 | ¹²⁹ I | 5.5 x 10 ⁻⁸ | 5.5 x 10 ⁻⁸ | 5.5 x 10 ⁻⁸ |
| ¹²⁹ Te | 5.5 x 10 ⁻³ | a | a | ²³⁸ U | 2.1 x 10 ⁻⁹ | 2.1 x 10 ⁻⁹ | 2.1 x 10 ⁻⁹ |
| ¹²⁷ Te | 3.8 x 10 ⁻² | 3.4 x 10 ⁻⁶ | a | ¹⁰⁷ Pd | 2.6 x 10 ⁻⁸ | 2.6 x 10 ⁻⁸ | 2.6 x 10 ⁻⁸ |
| ¹³⁴ Cs | 5.1 x 10 ⁻² | 1.3 x 10 ⁻² | 2.4 x 10 ⁻³ | ²³⁷ Np | 1.3 x 10 ⁻⁹ | 1.3 x 10 ⁻⁹ | 1.3 x 10 ⁻⁹ |
| ¹⁵¹ Sm | 4.4 x 10 ⁻³ | 4.3 x 10 ⁻³ | 4.1 x 10 ⁻³ | ¹⁵² Eu | 1.0 x 10 ⁻⁸ | 7.8 x 10 ⁻⁹ | 6.0 x 10 ⁻⁹ |
| ²³⁸ Pu | 3.8 x 10 ⁻⁵ | 3.7 x 10 ⁻⁵ | 3.5 x 10 ⁻⁵ | ²⁴² Pu | a | a | a |
| ²⁴¹ Pu | 8.1 x 10 ⁻⁶ | 6.7 x 10 ⁻⁶ | 5.4 x 10 ⁻⁶ | ¹⁵⁸ Tb | a | a | a |
| ²⁴⁴ Cm | 4.5 x 10 ⁻⁶ | 3.8 x 10 ⁻⁶ | 3.2 x 10 ⁻⁶ | ²³⁵ U | a | a | a |

a. Value <1 x 10⁻⁹.

TABLE IV-5

Chemical Composition of Reconstituted SRP
High-Level Waste

| <i>Constituent</i> | <i>Concentration</i> | |
|---------------------------------|----------------------|------------|
| | <i>Molar</i> | <i>g/L</i> |
| NaNO ₃ | 2.2 | 187 |
| NaNO ₂ | 1.1 | 76 |
| NaAl(OH) ₄ | 0.5 | 59 |
| NaOH | 0.75 | 30 |
| Na ₂ CO ₃ | 0.3 | 32 |
| Na ₂ SO ₄ | 0.3 | 43 |
| Fe(OH) ₃ | 0.07 | 7.5 |
| MnO ₂ | 0.02 | 1.7 |
| Hg(OH) ₂ | 0.002 | 0.5 |
| Other Solids | 0.13 ^a | 7.8 |

a. Assuming an average molecular weight of 60.

TABLE IV-6

Radionuclide Content of Reconstituted SRP
High-Level Waste (1985)

| <i>Radionuclide</i> | <i>Activity, Ci/gal</i> | <i>Total Activity, Ci</i> |
|--------------------------------------|-------------------------|---------------------------|
| ⁹⁰ Sr | 2.1 | 1.3 × 10 ⁸ |
| ¹³⁷ Cs | 2.2 | 1.3 × 10 ⁸ |
| ¹⁴⁷ Pm | 0.77 | 4.6 × 10 ⁷ |
| ¹⁴⁴ Ce- ¹⁴⁴ Pr | 0.19 | 1.1 × 10 ⁷ |
| ¹⁵¹ Sm | 0.07 | 4.2 × 10 ⁶ |
| ¹⁰⁶ Ru- ¹⁰⁶ Rh | 0.03 | 1.8 × 10 ⁶ |
| ²³⁸ Pu | 0.01 | 6.0 × 10 ⁵ |
| ²⁴¹ Am | 0.001 | 6.0 × 10 ⁴ |
| ²⁴⁴ Cm | 0.001 | 6.0 × 10 ⁴ |
| ²³⁹ Pu | 0.0004 | 2.4 × 10 ⁴ |

3. Differences Between Savannah River, Hanford, Idaho, and Commercial Wastes

High-level radioactive wastes generated at Hanford are similar to those at the Savannah River Plant in chemical and radionuclide composition. Hanford wastes are also processed to excess alkalinity and transferred to large underground storage tanks. However, the high-heat wastes at Hanford are not cooled like those at SRP; therefore, sludge in the SRP tanks remains more flocculent. Radiocesium and radiostrontium are being removed from the Hanford waste and stored in double-wall canisters as cesium chloride and strontium fluoride. Cesium and strontium removal operations are expected to be completed in the early 1980s.¹ Idaho National Engineering Laboratory (INEL) high-level waste composition varies greatly depending on the type of fuel being processed, the irradiation history of the fuel, and the length of time the fuel is stored before processing.² Unlike Savannah River and Hanford, INEL high-level liquid waste is initially stored as an acid solution and contains high fluoride concentrations. After a suitable decay period, the acidic waste is converted to a granular solid in a fluidized-bed calciner. For further details on the composition of INEL waste, see the *Final Environmental Statement, Waste Management Operations, Idaho National Engineering Laboratory* (Report ERDA-1536).³

The commercial high-level nuclear wastes at the Nuclear Fuel Services Plant at West Valley, New York are quite similar to the SRP alkaline wastes. However, the NFS wastes also include a small quantity of acid Thorex waste.

If reprocessing of nuclear power reactor fuels is ever resumed, the waste from the reprocessing plants will be similar to INEL high-level waste before calcining, except that it will contain less aluminum, zirconium, and fluorides, and a higher concentration of radionuclides, which in turn will generate more heat per unit volume. Wastes from nuclear reprocessing plants for power reactor fuel are described in *Alternatives for Managing Waste from Reactors and Post-Fission Operations in the LWR Fuel Cycle* (Report ERDA-76-43).²

B. TECHNOLOGY OF THREE MAJOR ALTERNATIVES FOR LONG-TERM WASTE MANAGEMENT*

1. Alternative 1 - Continue Storage in Tanks

Description

This alternative is a continuation of present high-level waste management practices at SRP and is therefore the "No Action" alternative under CEQ designations. However, since a considerable amount of positive action is required over a long time period to carry out this alternative, it is herein referred to as "Continued Present Action."

For purposes of calculating waste volumes, the DWD and this EIS have assumed SRP reactor operation ceases in 1987.** Under this assumption, the backlog of high-level waste to be managed will be stored in 25 tanks. Each tank would contain less than 1 million gallons, and would have a capacity of 1.3 million gallons. They would be the double-wall Type III design now being built at SRP. Use of heat-treated steels and stress relief after construction is expected to result in a service lifetime of at least 50 years for these tanks.

New tanks would be built as required by the observed condition of the tanks in service. Salt or sludge would be reconstituted to liquid by dissolving or slurring with water. This solution would be transferred to a new tank and evaporated to a damp salt cake or sludge as it was before transfer. The old tank would be cleaned and retired from service. The cycle of reconstitution to liquid, transfer to new tanks and evaporation, and retirement of old tanks would continue about every 50 years into the future. The process would cease when some future generation made a decision that some other disposal method would be more desirable, or that the radioactivity had decayed enough so that the tanks could be covered and abandoned.

The operations outlined above are described in detail in Reference 4.

* Other alternatives and reasons for their exclusion from coverage in this document are discussed in Section IX.

** The normal design of a plant for the remote handling of large quantities of radioactive materials provides for safe operation over an extended period of years. A waste solidification plant would thus be operable beyond the time necessary to work off a backlog determined by a 1987 shutdown. If operations were extended past 1987, increases in impacts such as consumption of materials and pre-emption of repository space would be expected to be roughly in proportion.

Status of Technology

This alternative is a continuation of operations currently performed at SRP on a routine basis, backed by about 25 years of experience. The technology for all necessary phases is therefore demonstrated full scale and in-hand. The lifetime of new, Type III stress-relieved tanks has not been demonstrated, but is projected from experience with other tanks and laboratory studies to be at least 50 years.

Research and Development Needed

No research and development effort is needed to implement this alternative at the current state-of-the-art level. However, improvements are being made in established methods of reconstituting waste, evaporation, level monitoring, tank surveillance, corrosion control, etc., as a result of small-scale, continuing research and development efforts. Current plans for these activities between now and the time that this alternative could be considered implemented (mid-1980) are discussed in detail in Reference 5.

2. Alternative 2, Subcase 1 — Immobilize* and Ship to a Federal Repository

The technology development program now under way and proposed for funding is oriented toward timely implementation of this alternative, which is illustrated in Figure IV-4.

Description

For all subcases of Alternative 2, salt solutions and slurried sludge are removed from the waste tanks in the two separations areas, F Area and H Area. These solutions are processed through a waste solidification plant which begins operation in 1988.

* Glass is used as the reference form in the analysis of Alternative 2 (all subcases). As stated in the foreword, the decision on waste form has not yet been made since another waste form will not be chosen unless it had less impact than glass, the analyses presented are bounding.

In the solidification plant, the sludge is washed and centrifuged free of residual salt. The salt solution is likewise filtered free of residual sludge and then passed through ion exchange columns to remove cesium and strontium, re-evaporated, and handled as described in Section IV.C. The sludge and ion exchange product are combined with SiO_2 , B_2O_3 , and other glass-making materials to form a matrix containing about 35% waste (25% on a waste oxide basis). The glass product is sealed in steel containers and shipped for offsite geologic disposal. For current reference purposes, the geologic disposal formation is assumed to be salt beds, but other geologic formations are also being considered.

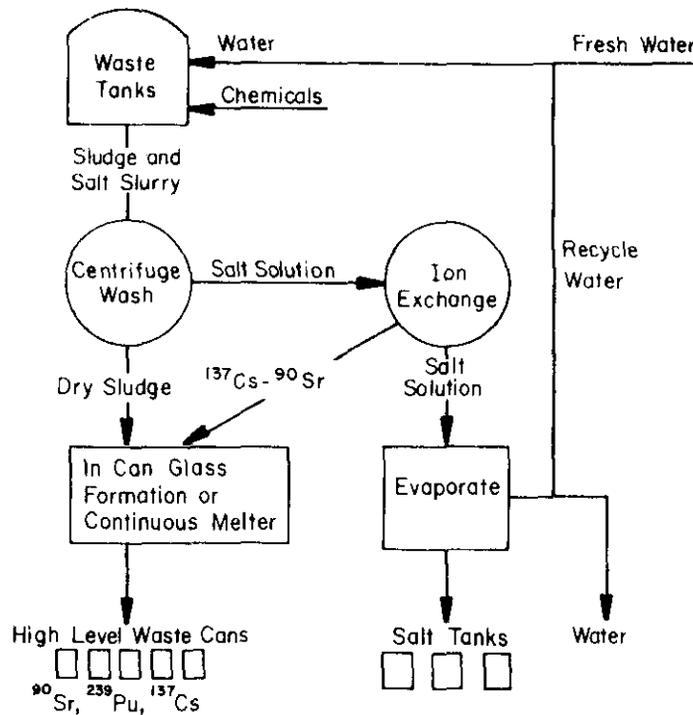


FIGURE IV-4. Conceptual Waste Solidification Process

A temporary storage facility is included to store up to two years of production of glass; the option of extended onsite storage is also open as discussed in Subcase 2.

Waste Removal and Tank Decontamination

Salt removal is accomplished by redissolving the salt in recirculated water and pumping the resultant solution from the tanks. Sludge removal is accomplished by slurring the sludge with special pumps and pumping the slurry from the tanks. Residual sludge is then removed by chemical cleaning with oxalic acid solutions.

By 1988 when the waste solidification plant starts up, all waste will be stored in modern, double-wall, 1.3 million gallon tanks. H Area will have ten tanks containing salt and liquid, one tank containing sludge, and five tanks containing liquid and sludge. F Area will have five tanks containing salt and liquid, three tanks containing liquid and sludge, and one tank containing sludge. Twenty-five will be in service by 1985.

Waste tanks are each fitted with low-pressure pumps for slurring and decontamination. Additionally, one mix tank in each area and the transfer tank in H Area are fitted with high-pressure pumps.

Waste transfer pumps (including installed spares) are required to move waste slurries from tank-to-tank and area-to-area. When possible, installed jets will be used to transfer salt solutions between tanks in the same area.

Additional equipment required for waste removal and tank decontamination includes:

- Slurring and transfer pumps
- Water recycle tanks – F and H Areas
- Oxalic acid solution tanks and pumps – F and H Areas
- Interarea water recycle transfer line
- Equipment for relocating pumps including a shielded cask on a flatbed vehicle
- Piping, valves, spray jets, spray rings, and other minor auxiliaries.

Sludge removal and tank cleanout have been demonstrated, but improved technology is currently being developed. Removal of aged high-heat sludge from a waste tank retired in service,

but still containing a sludge heel, was successfully demonstrated during FY 1979. Recirculated waste supernate was used as the slurring medium.⁶

Salt Decontamination

The waste solidification plant for processing the dissolved-salt slurried-sludge mixture is under conceptual design with a new canyon-type building located just outside H Area. Present design calls for separate streams of salt solution and thick sludge to be transferred to the processing building. The salt concentrate must be further clarified of sludge fines before ion exchange processing. Sand filtration has been used in tests, and it, along with agglomeration and etched disk filtration, is being evaluated in further tests.

The principal radionuclide, ¹³⁷Cs, is removed from the dissolved salt by sorption on a phenolsulfonic ion exchange resin such as *Duolite** ARC-359. ¹⁰⁶Ru would not be removed, but with its 1-year half-life, will decay to innocuous levels in about 10 years. The small amount of ⁹⁰Sr (and some of the other lower concentration lanthanides and actinides) in the dissolved salt would be removed by an additional stage of ion exchange using a chelating resin. With this step added, principal residual activities in salt would be 1.5×10^3 nCi/g ¹³⁷Cs, 30 nCi/g ⁹⁰Sr, and <2 nCi/g Pu.

Equipment to perform these processes remotely at large scale is being developed and demonstrated in a semiworks mockup with nonradioactive synthetic wastes.

The heart of the waste solidification process is the incorporation of the radionuclides into a high-integrity, low-leachability matrix. Glass is being developed as the matrix in the SRP studies, but, as discussed in Section IV.D, a number of other matrices are being developed in companion programs at other sites. The options are still being preserved in the SRP design program to use any of the possible immobilization matrices. Current development of the glass process is based on the light water reactor (LWR) waste vitrification process being developed by Battelle-Pacific Northwest Laboratories (PNL). As the first step in incorporating the waste into glass, the washed sludge and ion exchange eluate are combined and converted to dry powder in a spray claciner. The powder flows by gravity with glass frit into a continuous, Joule-heated electric furnace. Molten glass is periodically poured into steel canisters. After cooling, lids are welded on the canisters, which are then

* Registered trademark of Diamond Shamrock Chemical Company.

tested, leak-checked and decontaminated. Each container holds 165 gallons of glass product with about 59 kilocuries of ^{90}Sr and ^{137}Cs . The heat output of each container is about 290 watts. Off-gases from both calciner and glass furnace are quenched, absorbed, and scrubbed before being released; scrub solutions are back-cycled to the waste feed.

The decontaminated salt solution is evaporated in two stages of bent tube evaporators as it is returned to existing waste tanks. The evaporator overheads are recycled for dissolution of more salt.

Status of Technology

Research and development to date have included a waste tank sampling program to provide the waste characteristics described previously in this section. Sludge and supernate have been separated on a small scale for both simulated waste and actual waste by centrifugation and filtration.⁶ Testing of prototype wiped-film evaporators with synthetic waste began in FY-1977.⁵ Several glass formulations have been prepared and evaluated in shielded cells using actual waste and the preferred formulation selected.⁷ Compatibility of different container materials has been evaluated for the different waste forms.⁸ Actual supernate has been used to demonstrate supernate decontamination at laboratory scale.⁹ Engineering and cost studies have been performed for the complete process.¹⁰ A research and development program in cooperation with Pacific Northwest Laboratories in Richland, Washington is continuing to evaluate the calcining and glass melting steps on a larger scale.

Research and Development Needed

Further research and development activities are planned in the areas of sludge removal, waste tank cleaning, calcining, deionization, glass melting, and others.

Still other research and development programs are devoted to alternatives to the reference processes. Besides those studies aimed at alternatives other than glass, alternative glass process studies are investigating in-can melting of glass, direct liquid fed melters, and a variety of other options.

The work on geologic repositories is being developed under a separate DOE program administered under the Office of Nuclear Waste Isolation (ONWI) and will be covered in a separate series

of EISs. However, the SRP R&D program involves many interactions with the ONWI program. These interactions include determinations of waste form integrity under repository conditions, development of canisters and engineered barriers for the waste forms, and risk analyses of the different waste forms under repository conditions.

3. Alternative 2, Subcase 2 — Immobilize and Store in Surface Facility at SRP

Description

The processing steps of removal from tanks through vitrification are the same as those described for Subcase 1. Canned glass or other waste forms are stored in a reinforced concrete structure designed to withstand earthquakes, tornadoes, and missiles. This facility provides for natural-draft cooling of the individual containers, and is connected to the waste solidification facility by a tunnel. Shielded equipment places the cans in the storage position. Provision is made to recycle damaged or suspect containers to the canning facility. A possible alternative to the reinforced concrete structure is a water basin. Engineering and cost studies for these facilities were based on Reference 10.

Status of Technology

The status of technology is the same as that for Subcase 1, with the addition of the work that has been done on the air-cooled surface storage vault. Most of this work involves the conceptual design and is documented in Reference 10 for the SRP facility.

Research and Development Needed

Research and development needed by Alternative 2, Subcase 2 is the same as that for Subcase 1, except that the items related to an offsite geologic repository would not be needed. The air-cooled surface storage facility would be built using conventional materials and construction techniques, and would require comparatively little new research and development beyond the site selection activities.

4. Alternative 2, Subcase 3 -- Immobilize and Dispose of in an SRP Bedrock Cavern

Description

The processing steps of waste removal from tanks through vitrification are the same as those for Subcase 1. The glass or other immobilized product is disposed of in a bedrock storage cavern below the Savannah River Plant site instead of in an offsite geologic storage facility. Previous studies¹¹ concluded that a cavern 1500 ft below the surface in the Triassic formation would be best. The head house and main access shaft for such a facility are tentatively located about eight miles from the separations area (H Area) in the southeastern one-third of the plantsite.

The waste forms are transferred in a cask to the head house where they are removed from the cask and lowered through the access shaft to tunnels in the Triassic rock. Specially designed machines transport the can to the storage position in the tunnel.

During the period of emplacement in the bedrock, cavern ventilation is provided and water inleakage is pumped out. After the tunnels are filled, the access and ventilation shafts are sealed and, in time, presumably would fill by seepage of water from the metamorphic rocks. After this filling, retrievability would depend on the integrity of the waste product and waste canisters and the ability to pump out the water. Retrievability could be extended beyond the cavern filling period if water pumping and surveillance were continued.

Status of Technology

The status of technology for Alternative 2, Subcase 3 is the same as that for Subcase 1 through the vitrification step. Conceptual design studies have been made for a bedrock cavern under the SRP site, and extensive drilling of test wells was done to establish the overall characteristics of the underlying rock. This preliminary work indicated a satisfactory site probably exists in a Triassic basin about 8 miles from the present separations areas, and about 1500 ft below the surface. An extensive description of the conceptual design and the geologic investigations carried out before work ceased on this concept in 1972 is given in Reference 11.

Research and Development Needed

The research and development needed for Alternative 2, Subcase 3 is the same as that for Subcase 1, except that the items related to an offsite geologic repository would not be needed. Instead, an extensive research and development effort would be required at SRP, with the objective of ensuring a high degree of confidence in the physical integrity of the bedrock cavern. This work would require more test drilling and construction of an exploratory shaft and tunnels. The same types of parameters would be measured as in Subcase 1. Particular emphasis would be placed on studying possible pathways to the Tuscaloosa aquifer, which lies above the potential bedrock cavern site. These requirements are discussed more fully in Reference 11, and would probably result in at least 10 years of increased time for implementation compared to other alternatives.

No development work oriented toward a bedrock cavern at SRP is under way, nor is any proposed for funding.

5. Alternative 3 – Dispose of Liquid Waste in an SRP Bedrock Cavern

Description

Present waste would be reconstituted to liquid as described for Alternative 2, Subcase 1, but with the salt and sludge streams combined. The waste slurry would be pumped about 8 miles through a heavily constructed double transfer line to a bedrock cavern. The cavern would be similar to that described for Alternative 2, Subcase 3, except that it would have a volume of about 17 million cubic feet to provide extra space for radiolytic gas, water inleakage, and rock creep (this is about 3 times the size of the cavern required for Alternative 2, Subcase 3).

Status of Technology

Reconstitution of present waste to liquid and transfer to the site of the bedrock cavern are similar to activities that are performed routinely in present waste management operations and would require no new technology development. Mining of the bedrock cavern is also within the capabilities of present-day routine mining.

Research and Development Needed

The research and development efforts for this alternative would be directed toward ensuring the integrity of the bedrock cavern, as described for Alternative 2 - Subcase 3, and in Reference 11.

This work is not now under way, and it is not currently proposed for funding. Furthermore, the U.S. Environmental Protection Agency in their comment letter on the draft of this EIS, has ruled this alternative to be Environmentally Unsatisfactory.

C. ALTERNATIVES FOR DECONTAMINATED SALT STORAGE

1. Description of Decontaminated Salt

The alternative that processes the high-level waste into an immobilization matrix also produces decontaminated salt. The dissolved salt removed from waste tanks is processed through a two-step ion exchange process, one to remove cesium and the other to remove strontium plus residual quantities of other lanthanides and actinides. The radionuclides eluted from the ion exchange columns are incorporated into the glass matrix.

Immediately after processing, the salt contains less than 1% of the radioactivity in the high-level waste. ^{106}Ru will be the predominant radionuclide in the salt. Its concentration depends on the age of the waste after the reactor irradiation producing the ^{106}Ru . After 10 years, this relatively short-lived radionuclide is reduced by a factor of one thousand, and after 20 years, by a factor of one million. The radioactivity in the salt after 10 years is shown in Table IV-7. In addition to the reference decontamination factors given in Table IV-7, the R&D program is also investigating the possibility of essentially complete decontamination of the waste after ^{106}Ru decay (^{106}Ru removal is also being investigated, but does not currently appear economically attractive). This extra decontamination might be performed in a second pass through the immobilization plant using different ion exchange resins if necessary, but the same equipment. It might also be accomplished with new, lightly shielded and relatively inexpensive equipment at the salt storage tanks.

After the dissolved salt solution is substantially decontaminated by ion exchange, it is evaporated in bent-tube and/or wiped-film evaporators and the concentrate is cooled to crystallized salt. The principal chemical composition of the salt, excluding any residual water, is shown in Table IV-8.¹²

TABLE IV-7

Radionuclide Content of Decontaminated Salt
(10-year-old waste)

| <i>Radionuclide</i> | <i>Concentration (nCi/g)</i> | |
|-----------------------------------|------------------------------|----------------------------|
| | <i>Chemically Measured</i> | <i>Computer-Calculated</i> |
| ³ H | NA ^c | 57 |
| ⁶⁰ Co | NA ^c | 390 |
| ⁹⁰ Sr-Y ^a | 2 | 9 |
| ⁹⁹ Tc | 125 | 220 |
| ¹⁰⁶ Ru-Rh ^b | 287,000 | 100,000 |
| ¹²⁹ I | NA ^c | 0.04 |
| ¹³⁷ Cs-Ba ^a | 100 | 480 |
| ¹⁴⁴ Ce-Pr ^b | 109 ^d | 220 ^d |
| ¹⁴⁷ Pm ^b | 100 ^d | 5200 ^d |
| ¹⁵¹ Sm | <10 ^d | 116 ^d |
| ¹⁵⁴ Eu | 71 ^d | 510 ^d |
| ²³⁸ Pu ^a | 9 | 0.9 |
| ²³⁹ Pu ^a | 0.3 | 0.02 |
| ²⁴⁰ Pu ^a | 0.3 | 0.02 |
| ²⁴¹ Pu ^a | 2 | 3.5 |
| ²⁴¹ Am ^a | 0.5 | 0.03 |

- a. With decontamination factors assumed Cs 10⁴, Sr 10³, actinides 10² (165 for computer-calculated concentrations).
- b. Decay of short-lived radionuclide may contribute to differences in computer-calculated and chemically measured concentrations.
- c. Not analyzed.
- d. Concentrations of rare-earth fission products should be reduced by a factor of 10² (165) during decontamination operations.

TABLE IV-8

Chemical Composition of Decontaminated,
Crystallized Salt

| <i>Component</i> | <i>Weight Fraction</i> |
|---------------------------------|------------------------|
| NaNO ₃ | 0.458 |
| NaNO ₂ | 0.186 |
| NaOH | 0.073 |
| NaAlO ₂ | 0.100 |
| NaCO ₃ | 0.078 |
| Na ₂ SO ₄ | 0.104 |

(Note that the nitrate fraction decreases and the nitrite fraction increases during the early years of storage.)

Research and development have not progressed to the extent that the concentration of mercury in the decontaminated salt can be determined precisely; however, the concentration is expected to be less than 4×10^{-4} grams of mercury per gram of salt. The total amount of Hg in the 16.3 million gallons (~120,000 tons) of salt would then be less than 60 tons.

2. Alternative Storage Modes

Store in Tanks at SRP

The decontaminated salt solution is transferred to tanks outside the canyon-type solidification facility and processed through evaporators. The concentrate is transferred to decontaminated double-wall carbon steel waste tanks encased in reinforced concrete (this is the current design, or Type III, tank). The steel tanks have an expected life of 50 to 100 years, and the 2.5-ft-thick concrete encasements have an expected life of several hundred years. The concentrate is cooled to form crystallized salt. If all the solution does not crystallize when cooled, the supernate is recycled for further concentration until it does crystallize.

The tanks are monitored at the same level as the current practice for SRP waste tanks. After one hundred years when the residual ⁹⁰Sr and ¹³⁷Cs in the salt have been reduced by a factor of 10 due to radioactive decay, the access ports through the tank covers will be plugged and sealed. Other protective provisions include a confinement barrier over the tanks, such as reinforced

concrete slab several feet thick with embedded durable warning signs, to prevent accidental intrusion and to obstruct malicious entry into the storage tanks. A 25-ft-thick earthen cover, faced with rock, is placed over the concrete slab to provide protection from surface conditions and to provide a landmark that will not be lost by weathering over the centuries. At the perimeter of the earthen mound, new monitoring wells are installed to allow continued surveillance if required.

Can and Store in an Onsite Surface Vault

The decontaminated salt solution is evaporated to form crystallized salt. Four cells in the canyon-type solidification facility are allotted to evaporate the salt solution and can the crystallized salt; two cells are allotted to evaporate the solution and can the salt; and two cells are allotted to decontaminate and inspect the canisters. The metal canisters are sealed by welding. After decontamination and inspection, the canisters are transferred to a surface storage vault. Because of the low radionuclide content of the salt, the canisters do not require forced cooling in the storage vault.

Can and Ship to an Offsite Federal Repository

The decontaminated salt solution is evaporated and placed in canisters identical to those proposed for use in a surface storage vault at SRP. The canisters are shipped to an offsite Federal repository for disposal.

Other Options After ^{106}Ru Decay

After ^{106}Ru decay, and particularly if a second stage of decontamination is used, the salt can be expected to be at a low enough activity that it can be treated essentially as a chemical, rather than a low-level radioactive waste. Possible options then available include shallow land burial in a dry location, sea disposal, and return of the material to commerce.

D. ALTERNATIVE WASTE IMMOBILIZATION FORMS

Disposal Alternative 2 calls for immobilizing SRP high-level waste in a high integrity form before placing it in a Federal Geologic Repository (Subcase 1), in a Surface Facility at SRP (Subcase 2), or in a Bedrock Cavern at SRP (Subcase 3). Borosilicate glass was selected in 1977 as the reference form for immobilization of SRP high-level waste, and a major effort is currently underway to develop the required technology. In addition DOE is investigating a number of alternative waste forms. A preliminary analysis of the waste forms will be completed in FY 1980. Forms that have potential superior product performance or process characteristics to those of glass will then be selected for more detailed review. Conceptual processes will be carefully defined for each selected waste form. These processes will be evaluated to provide improved assessments of performance attributes and will provide the basis for better quality cost estimates. Sufficient data is expected to be available in the form of regulatory criteria and from the waste form development and characterization program to provide a basis for a detailed systems assessment in FY 1983. The final waste form for immobilization of SRP high-level waste will be selected by the end of FY 1983 based on the results of the systems assessment. (See Figure IV-5 for overall evaluation schedule.)

1. Waste form Requirements

The high-level waste immobilization form must meet a number of different requirements at different stages of the waste disposal process; it is essential that it be considered in such a total system context rather than merely in terms of any single factor such as long-term leachability. These requirements include, by disposal stage:

a. Processing

- o The waste form must be produced by a safe, practical process at acceptable cost.
- o The waste form must be flexible enough to accept reasonable variations in waste composition and process conditions.

- The waste form must be certifiable in terms of process quality control and quality assurance testing.
- The waste form should desirably be amenable to second-generation improvements.

b. Interim Storage

- The waste form must be resistant to handling and short-term corrosion.
- The chance of radionuclide dispersal must be low on coolant loss or sabotage.

c. Transportation

- The waste form must be resistant to transportation accidents, including impact, short-term leaching, and hot fires.
- The waste form must be resistant to sabotage.

d. Repository Emplacement

- The waste form must meet repository handling requirements in regard to structural integrity, surface contamination, fire resistance, dimensions, weight, etc.
- The waste form must meet repository retrievability requirements. The requirements are not yet fixed but can be expected to be between 20 and 50 years.

e. Repository Storage

- The primary requirements are for low leachability under repository conditions assuming both static water (normal storage) and flowing water (accidents), and for good compatibility between the waste form and the host geologic medium.
- The secondary requirement is resistance to dispersal in accidental or deliberate intrusion.

The final arbitrator of these requirements is the projected health risk to man from exposure to radionuclides released by the waste form. On this basis, the waste form represents merely the final barrier in a multibarrier system to reduce the health risk. In no case is the waste form the primary barrier. Thus, for processing, the primary barrier is the processing building and

the engineered containment of the processing equipment; for interim storage it is the storage basins and the waste canister; for transportation it is the waste canister and the shipping cask (already well developed for spent fuel handling); for repository emplacement, it is the canister, the repository, and the emplacement equipment; while for repository storage it is the canister, the repository itself, plus any engineered features such as overpacks. Despite the secondary nature of the barrier afforded by the waste form, the multibarrier concept still calls for the waste form to provide independent protection so as to maintain an acceptably small risk level even if any of the primary barriers should fail.

2. Borosilicate Glass Waste Forms

Borosilicate glass has to date been the waste form of most interest in the high-level waste disposal programs both in the U.S. and abroad.¹³ It is now in commercial use in the European waste management programs. (See Table IV-9 for list of foreign HLW form programs.) One of the advantages of borosilicate glass is that it can accommodate a large variety of glass formers and waste compositions. The waste glass can thus be tailored to the particular waste composition and to the particular processing equipment and conditions. Table IV-10 gives a sample composition for one SRP borosilicate glass form.

The advantages of borosilicate glass for high-level waste immobilization include the following:

- Glass technology is well developed and uses simple, easily available materials.
- Extensive technology has been developed at Pacific Northwest Laboratories and elsewhere specifically for fabricating high-level waste forms.
- Borosilicate glass will accommodate essentially all the waste radionuclides except the noble gases, although a few of the more volatile like Cs, I, and Ru may have to be partially recycled from an off-gas system during glass formation. Borosilicate glass will also accommodate essentially all the nonradioactive elements in the SRP high-level waste sludge.
- The glass will accommodate relatively high waste loadings (~28 wt % total waste oxides) to produce a relatively high density product (~2.7 g/cm³).
- Glass properties are not critically dependent on waste composition, glass former compositions, or processing conditions.

TABLE IV-9

International HLW Immobilization Status

| <i>Nation</i> | <i>Process</i> | <i>Status/Major Milestone</i> |
|---------------|------------------------------------|--|
| France | Borosilicate Glass - "AVM" | 0.5 ton/day hot pilot plant startup 1978-1979 Production plant startup 1982-1983 |
| Germany | Borosilicate Glass | "VERA" 0.5 ton/day cold pilot plant operation considering French "AVM" process for licensing |
| Eurochem | Borosilicate Glass Metal Matrix | French "AVM" selected for production plant VITRAMET - LOTES) Pilot Plant VITRAMET - PAMELA) 1981-1982 |
| England | Borosilicate Glass | "FINGAL-HARVEST" production plant 1990 French "AVM" under consideration |
| Russia | Phosphate Glass | Cold pilot-plant work in progress |
| India | Borosilicate Glass | 0.1 ton/day hot plant startup 1979-1980 |
| Japan | Glass or Ceramic | Hot demonstration plant 1986 |
| Sweden | Ceramic | Laboratory studies in progress |

TABLE IV-10

Composition of Typical SRP Borosilicate Glass^{a, b}

| <i>Calcine Composition</i> | | <i>Frit Composition</i> | |
|---------------------------------|-----------|-------------------------------|-----------|
| Fe ₂ O ₃ | 42.0 wt % | SiO ₂ | 52.5 wt % |
| Al ₂ O ₃ | 8.5 | B ₂ O ₃ | 10.0 |
| MnO ₂ | 11.8 | Na ₂ O | 18.5 |
| U ₃ O ₈ | 3.9 | Li ₂ O | 4.0 |
| NiO | 5.2 | CaO | 4.0 |
| SiO ₂ | 3.8 | TiO ₂ | 10.0 |
| Na ₂ O | 4.7 | | |
| Zeolite | 8.8 | | |
| NaNO ₃ | 2.6 | | |
| NaNO ₂ | 0.2 | | |
| NaAlO ₂ | 0.2 | | |
| NaOH | 3.9 | | |
| Na ₂ SO ₄ | 1.3 | | |

a. Glass will contain 28 wt % calcine.

b. Average density of glass will be 2.7 g/cm³.

- The glass may be cast in large monoliths with a relatively small degree of cracking (based on present measurements, cracking approximately doubles the monolith surface area).
- The glass monoliths are compatible with relatively inexpensive and conventional stainless steel or carbon steel canisters.
- The canistered glass monoliths are structurally strong, have good impact resistance, and a high heat capacity. They require very large energy inputs to fracture them into airborne powder or to volatilize them.
- The glass has good resistance to radiation damage and to internal helium buildup (from radioactive alpha decay in the contained waste). Equivalent exposures of up to 10^6 years in radioactively spiked samples have shown essentially no radiation damage effects. Stored energy from the exposure was less than 35 cal/g.
- The glass is nonflammable and emits essentially no gases or volatile radionuclides at temperatures under 700°C . The canister may be kept contamination-free on the outside.
- The glass has good resistance to water leaching. At their expected surface temperature of $\sim 100^{\circ}\text{C}$ (a maximum of 130°C if five-year-old waste is ever processed for immediate disposal), the SRP glass waste forms have measured leachabilities of about 10^{-6} to 10^{-7} g/cm²/day.
- The glass has not been observed to devitrify at temperatures under $500\text{-}600^{\circ}\text{C}$, and even when it is devitrified, its leachability at expected repository conditions still remains in the 10^{-6} range.

The possible objections which have been raised against glass as a high-level waste immobilization form, have to do primarily with its behavior during high temperature leaching tests. At temperatures of 350°C and high pressures (well above the $100\text{-}130^{\circ}\text{C}$ surface temperature expected for glass monoliths made from the SRP wastes), glass leaches relatively rapid ($\sim 10^{-2}$ g/cm²/day) in distilled water and shows extensive substitution and devitrification in brine. A possible interpretation of these tests is that they represent a form of accelerated testing which demonstrates inherent thermodynamic instability for glass as compared with some of the crystalline waste forms. Interpretation of any of the leaching experiments is complicated by the fact that glass leaching is a complex multistep process involving gel formation by water penetration, interstitial element diffusion, element replacement, devitrification, and glass structure dissolution. This complicated

series of mechanisms in turn complicates the extrapolation from the relatively brief leaching tests to the long-term repository behavior.

3. Other Waste Immobilization Forms

Table IV-11 compares a number of possible high-level waste immobilization forms in terms of some processing and performance parameters. In many cases the judgments listed in the table are both qualitative and preliminary pending execution of the research programs to be described in the next section, but they do indicate the range of parameters to be expected.

The first three waste forms listed include calcine in which the waste is fired to a mixture of oxides at 300-700°C, rich clay in which the waste is solidified by mixing with clay to absorb water, and normal concrete in which the waste is set to a solid in cement. These forms are the primary choices for in-place immobilization of the wastes. They use available technology, they are marginal-to-good in leach resistance, but they offer little intrinsic resistance to transport accidents, thus putting almost all the reliance on the shipping cask.

The next forms listed include hot pressed concrete in which interconnected voids and excess water have been eliminated from the normal concrete, pelletized calcine in which the calcine has been agglomerated and some of its water solubility has been removed by firing the waste with various additives in the calcining process, glass in which advanced high temperature glasses might be considered in addition to the current waste glasses, and clay ceramics in which the waste-clay mixtures are fired to semiconventional ceramics. These forms are viewed as the current choices for the near-term waste immobilization plant at Savannah River. Current available evidence indicates glass to be the best of these forms.

The next three forms listed include supercalcine in which extensive additives are incorporated in the calcine mixture with the intention of producing an assemblage of highly stable, highly leach-resistant (mainly) silicate minerals after firing, SYNROC in which firing or hot pressing is used to produce a similar series of titanate minerals, glass ceramics in which a waste glass is deliberately partially devitrified under controlled conditions again with the intent of producing highly stable forms. Finally, the last three forms listed are representative of possible composite waste forms. They include metal matrices in which pellets of glass, supercalcine, or other waste forms are incorporated in a metal binder, multibarrier forms in which the individual waste particles are coated with carbon, Al_2O_3 , or other impervious materials before metal encapsulations, and cermets in which very small waste particles are formed *in situ* in the metal matrix.

TABLE IV-11

High-Level Nuclear Waste Immobilization Forms – Properties Comparison

| Waste Form | Devel. Status | Process Complexity | Process Flexibility | Waste Loading | Dispersion Impact Resis. | Long-Term Stability | Fire Resistance | Leachability | |
|-------------------------------------|---------------|--------------------|---------------------|---------------|--------------------------|---------------------|-----------------|--------------|--------|
| | | | | | | | | 100° C | 350° C |
| Calcine | Available | Low | Excellent | High | Very Low | High | Poor | Poor | Poor |
| Rich Clay | Available | Low | Excellent | Low | Low | ? | Poor | Medium | Poor |
| Normal Concrete | Available | Medium | Excellent | Medium | Medium | Medium | Poor | Medium | Poor |
| Hot, Pressed Concrete | 5 years | High | Excellent | Medium | High | Medium | Medium | Good | Poor |
| Pelletized Calcine | 5 years | High | Excellent | Medium | Medium | Medium | Medium | Good | Poor |
| Glass | Available | High | Excellent | Medium | High | High | Excellent | Excellent | Poor |
| Clay Ceramic | 5 years | High | Poor | Medium | High | Medium | Medium | Good | Poor |
| Supercalcine | 15 years | Very High | Poor | High | Very High | High ? | Best | Best | Poor |
| Synroc | 15 years | High | Poor | Very Low | Very High | High ? | Best | Best | Good |
| Glass Ceramic | 15 years | Very High | Poor | Medium | High | High | Excellent | Excellent | Poor |
| Pellet in Metal Matrix | 5 years | Very High | Good | Low | Very High | High | Poor | Excellent | Poor |
| Coated Supercalcine in Metal Matrix | 15 years | Highest | Poor | Medium | Very High | High ? | Excellent | Best | Poor |
| Cermet | 10 years | Highest | Poor | Medium | High | High | Excellent | Excellent | Poor |

Most Attractive
Intermediate
Least Attractive

IV-32

These six forms (and other closely related forms) are the primary candidates for advanced waste form development. None of them, with the probable exception of glass marbles in a metal matrix, is available for use now or is even well characterized. Thus, the excellent properties listed for these forms in Table IV-11 are to some extent tautological in that the development simply aims at achieving these properties. However, the possibility of such an achievement is in most cases supported by limited experimental data.

4. Waste Form Development Program

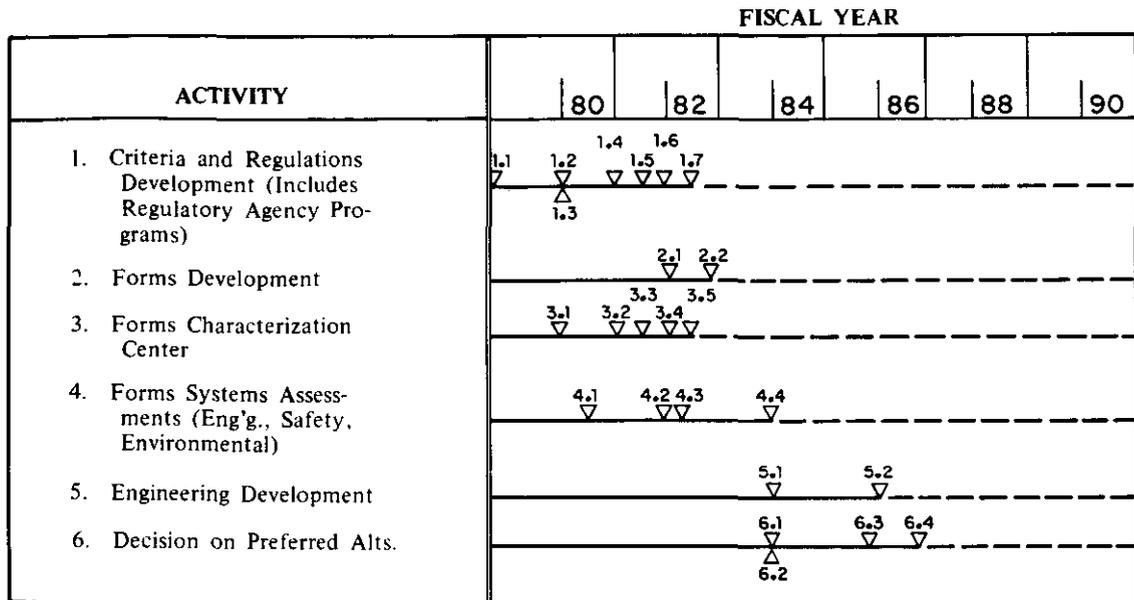
Essentially all the waste forms listed in Table IV-11 are under active development. The development programs are widely dispersed through the waste sites, the DOE national laboratories, industrial laboratories, and universities in order to secure the widest possible input. They are summarized in Table IV-12 and discussed briefly below for each of the major types of forms. Figure IV-5 shows the schedule and key milestones.

Calcine — Calcine waste form development is largely centered at the Idaho Chemical Processing Plant (ICPP),¹⁴ where a long-term program has been pursued to calcine all the plant high-level waste for interim or permanent storage. Current ICPP studies on calcine waste forms are primarily concentrated on pelletizing the existing calcines either for direct disposal or for incorporation in a matrix system. Work on calcines is also under way in the super-calcine program and, as a calcine intermediate, on the borosilicate glass programs at PNL and SRL.

Rich Clay — Work on the rich clay and related clay solidification forms is largely being conducted at Hanford as a means for in-tank solidification of the Hanford wastes.

Polymers — Synthetic and natural (bitumin) polymers are in wide use for immobilizing low-level and transuranic wastes,¹⁵ and Brookhaven National Laboratory (BNL) has done some work on polymers for high-level waste forms. However, the radiation and long-term stability of the HLW polymers is marginal. More important they are flammable, and in the case of the defense wastes which contain nitrate and permanganate as oxidizing agents, potentially explosive. Hence, no work is under way on polymer high-level waste forms.

Normal Concrete — A great deal of work has been performed on normal concrete high-level waste forms.^{16,17,18} Major efforts have been carried out at Brookhaven National Laboratory, Oak Ridge National Laboratory, Penn State University, and Savannah River Laboratory. However, most current work on concrete waste



- | | | |
|---|---|------------------------------------|
| 1.1 EPA Draft Gen. Criteria | 3.1 Established | 5.1 Glass Forms |
| 1.2 Initiate NRC Staff Tech Position Papers | 3.2 Fully Operational | 5.2 Alternatives |
| 1.3 Preliminary ONWI Criteria | 3.3 Testing Methods for Waste Forms Developed | 6.1 DWPF Final Form |
| 1.4 EPA Draft Tech Criteria | 3.4 Mat'ls Handbook Initiated | 6.2 <i>Second Generation Forms</i> |
| 1.5 NRC Draft Regulations (10CFR-60) | 3.5 Methods for Barrier Mat'ls | 6.3 Alts. for ICPP, Hanford |
| 1.6 Final ONWI Criteria | 4.1 Preliminary Glass | 6.4 Form for Comm. Waste |
| 1.7 NRC Final Regulations | 4.2 Preliminary Alts. | |
| 2.1 Glass Formulations Selected | 4.3 Final Glass | |
| 2.2 Alternatives Selected | 4.4 Final Alternatives | |

FIGURE IV-5. National HLW Long-Term Management Program Waste Form Selection

TABLE IV-12

Alternate Waste Form Development

| <i>Current Programs</i> | <i>Location</i> |
|-------------------------------------|-----------------|
| Supercalcine, Coatings and Matrices | PNL |
| Sintered and Coated Calcine | ICPP |
| Glass Marble - Metal Matrices | ANL |
| Concrete (FUETAP) | ORNL |
| Cermet | ORNL |
| SYNROC | LLL |
| Sol-Gel | ORNL |

forms is concentrated on in-place applications such as the ORNL shale hydrofracturing with grout, on the newer higher integrity concretes assumed below, or on low-level waste applications.

Hot-Pressed Concrete — Development of hot-pressed concrete waste forms is being pursued primarily at Oak Ridge National Laboratory under their FUETAP (Formed Under Elevated Temperature and Pressure) program.¹⁹ Pennsylvania State University has also developed hot-pressed concrete forms.²⁰

Pelletized Calcine — Pelletized calcines are being developed primarily in the ICPP calcine programs.²¹

Borosilicate Glass - Borosilicate glass is the most developed form. The U.S. effort is primarily focused at Pacific Northwest Laboratories (PNL)²² and at Savannah River Laboratory (SRL).^{6, 23} Work on adapting the borosilicate glass to their particular waste forms is also under way at each of the waste sites. In addition, as listed in Table IV-9, most other nations have borosilicate glass waste programs.

Phosphate Glass — Phosphate glass has received considerable attention as a waste form. U.S. studies have been concentrated at BNL and PNL; a number of studies have also been carried out in Russia and Western Europe, particularly Germany. However, these studies uncovered major problems in devitrification (low temperatures of crystallization with a major increase in leachability) and in incompatibility with container materials. Hence, no U.S. study is currently under way on these glasses.

High-Silica Glasses — High-silica natural glasses (obsidians and tektites) are known to have persisted for long periods in both terrestrial and lunar environments. However, these glasses work at about 1600°C, temperature high enough to drive off most of the ruthenium and cesium radionuclides from the waste. Investigation²⁴ is being made of several proprietary processes for low-temperature formation of high-silica glasses containing high-level wastes.

Clay Ceramics — Adding aluminum silicate clays such as keolin or bentonite to the waste typically produces an insoluble cancrinite-type material. This material can be fired to a nepheline-like ceramic. Some consideration is being given to these materials in the Hanford program. However, most of the attention is focused on the more-advanced ceramic analogues of long-lasting natural minerals in the supercalcine and SYNROC programs considered below.

Supercalcine — Pennsylvania State University,^{25,26,27} working in cooperation with Pacific Northwest Laboratory, has added various silicate materials in the waste calcining process to produce synthetic analogues of natural silicate minerals which can be hot pressed or sintered to ceramic waste forms. Penn State is continuing this work in cooperation with PNL and the Rockwell International Company in one of the larger scale waste form development programs.

SYNROC — Prof. Ringwood at the Australian National University has developed²⁸ several assemblages of synthetic titanate minerals, which he calls SYNROC, as waste forms. As with the supercalcines, the SYNROC synthetic minerals are based on natural analogues that have persisted in nature for very long times and that can be sintered or hot-pressed to ceramic forms. Lawrence Livermore Laboratory is working with Prof. Ringwood on SYNROC development. In addition, SYNROC-type compositions are being looked at in a number of the other U.S. waste form programs on an exploratory basis.

Titanates, Niobates, Zirconates — Sandia Laboratories²⁹ have developed these materials as mineral ion exchangers and are pursuing a small program to determine the practicality of hot pressing or sintering them to waste forms. These materials are also being considered as engineered barriers around the waste forms.

Glass Ceramics — One form of glass ceramic can be made by sintering or hot pressing the mixture of waste and glass frit rather than melting it as in normal glass-making practice. The resulting lower temperature processes have some attraction in reducing radionuclide volatilization and chemical corrosion; they have received limited attention for the fluoride-containing wastes at ICPP.³⁰ The more common forms of glass ceramics are formed by controlled devitrification. PNL is pursuing a small program in this area in cooperation with a larger program at the Federal Republic of Germany.

Metal Matrices — Most of the waste forms discussed above such as calcines, concretes, glasses, ceramics, and artificial minerals can be formed in small sizes and dispersed in a metal matrix for better heat transfer, reduced frangibility, easier sampling, and additional leaching barriers. Low melting alloys such as Pb-Sb/Sn or Al-Si can be cast around the waste particles, while higher melting metals can be sintered around the particles at temperatures of about two-thirds their melting temperature. Metal matrix waste form work in the U.S. is primarily concentrated at Argonne National Laboratory³¹ and Pacific Northwest Laboratory,^{32,33} with smaller programs at the Idaho Chemical Processing Plant and elsewhere.

Multibarrier Forms -- More complex matrix waste forms can be made by coating the waste particles with impervious materials such as carbon, alumina, or silicon carbide before placing them in the matrix. Such coatings provide additional barriers against waste leaching and also allow the use of higher temperature matrix-forming processes by reducing radionuclide volatilization. Concretes, sintered ceramics, and other materials can be used rather than metal as the matrix, if desired, when coated particles are used. The primary U.S. effort on multibarrier forms has been performed at Battelle Memorial Institute in their Pacific Northwest and Columbus Laboratories.^{32, 33} Consideration is also being given to applying to the multibarrier forms the coated particle technology developed by General Atomics for their high temperature gas cooled reactors (HTGRs).

Cermets -- Cermet high-level waste forms are a particular matrix form in which very fine waste particles are dispersed in a metal matrix, usually by *in situ* precipitation. Oak Ridge National Laboratory^{34, 35} is developing a particular waste cermet in which the wastes (and additional metal formers) are dissolved in urea, and the metal formers are reduced from the solution to form a *Hastelloy**-like alloy containing finely dispersed nonmetal waste particles.

Fused Salt - This waste form is not currently under active development. The distinctive characteristic of the fused salt waste form is that no separation of salt and sludge is made, and the waste is not processed through ion exchange to a high activity fraction and a residual low activity fraction. Salt is dissolved from the tanks and sludge is suspended in salt solution. The solutions are blended and fed to a low temperature fusion plant. Water is removed in successive stages of evaporation and dehydration, using bent tube and wiped film evaporators and rotary melters. The water removed from the salt is recycled to the waste tanks for dissolution of additional salt.

Preliminary studies have shown that 1000-gallon containers can be used for fused salt without exceeding design centerline temperatures and transportation weight limits. Each container holds 1000 gallons of fused salt and contains 22.6 kilocuries of ⁹⁰Sr-¹³⁷Cs with a heat output of 130 watts.

Because the low temperature fusion process is simpler than partitioning waste and manufacturing glass or concrete, full-scale testing and implementation lead times would fall well within the schedules planned for previously described alternative plans.

Fused salt is a less expensive product than glass or concrete. Although the processing step of conversion to fused salt is about as expensive as vitrification or cementation, the salt decontamination step is eliminated completely. Somewhat higher container costs are incurred because of the larger volume of product, but the expense of returning decontaminated salt to tanks is eliminated.

Fused salt has risks similar to dry powder with regard to water intrusion. The risks from airborne particles is intermediate between dry powder and glass. Sabotage during processing carries a lower risk than that for other waste forms because there is no point in the processing operation where concentrated, fine particles are available.

Fused salt is not proposed for specific research and development activities because its costs and risks are intermediate between glass and the other major alternatives of continued tank farm operation or disposal of liquid waste in SRP bedrock. These product forms may not satisfy the desire of the general public regarding a high integrity waste product and the role the form would play in the multiple barrier concept. Most of the processes needed for a fused salt product would, however, be investigated in the course of an advanced form product development, so they are not precluded by the current program.

5. Canister and Engineered Barrier Programs

The waste forms are normally housed in a canister which provides a contamination-free handling surface, add mechanical strength, provides extra containment during shipping, handling, and interim storage, and also provides an additional barrier against repository waste leaching. This canister may in turn be surrounded by additional isolation materials, secondary canisters, and other engineered barriers. In many cases these engineered barriers are designed to provide radionuclide containment equivalent to that of the waste form itself. In the repositories they act to restrict and condition any flow of repository water to the waste form and similarly to restrict and condition any flow of leached radionuclides from the waste form to the repository.

Since the canisters and other engineered barriers form the interface between the waste form and the repository, they must be considered jointly by the waste form, transportation, and repository programs. In general, however, the waste forms program has primary responsibility for the initial canister and for any other engineered barriers added at the forms production plant; the transportation program has primary responsibility for engineered barriers in the shipping canisters; and the repository

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program has primary responsibility for any engineered barriers added to the repository.

Each of the waste form programs includes a program for the primary canister to contain that form. More general canisterization and engineered barrier programs are also being conducted under the high-level waste management programs at Oak Ridge National Laboratory, Pacific Northwest Laboratory, and Savannah River Laboratory. Other types of engineered barrier studies are under way at Hanford and the ICPP for the options of in-tank immobilization of the wastes at these sites.

6. High-Level Waste Forms Characterization

High-level waste forms characterization serves two different purposes. First, it provides a basis for waste forms selection by comparison of the properties of the different waste forms with each other and with the waste form criteria and regulations. Second, once the waste form has been selected, it provides a means of quality assurance that the waste forms have been properly produced and that they meet the required standards. The first purpose is largely served by the properties tests of each form, and the second purpose is largely served by the quality assurance tests in each production program. However, a limited external characterization program is also necessary outside the direct development and production programs to make sure that all the candidate forms are evaluated on a uniform basis and that the quality assurance tests do indeed meet the regulatory requirements.

A waste characterization center is being established at Battelle Pacific Northwest Laboratory, (A DOE national laboratory is specified because of the need to handle large amounts of radioactivity in some of the tests.) The characterization center will develop required testing procedures and issue them for distribution, determine the relation between these procedures and any applicable regulations, issue reference data on each of the candidate waste forms, and verify data collected in the development and production programs.

In order to provide quality assurance on the characterization center data and to secure system-wide concurrence, the characterization center results will be issued through a materials review board composed of data users, independent experts, and a representative of a separate certification laboratory. The latter will be established in the separate waste repositories reporting chain and will provide independent quality assurance testing of the characterization center results.

The final components in the characterization program are the scientific laboratories. The purpose of these laboratories is to relate the observed properties of the waste forms to the fundamental processes underlying these properties. In most cases the properties measurements are made over a span of a few hours -- at most a few years -- and then extrapolated to hundreds or thousands of years for decision-making between waste forms. Since many of the waste form properties are nonlinear with time, such extrapolations can be made only in terms of the fundamental mechanisms underlying the measured properties.

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