QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
WASTE FIXATION PROGRAM
APRIL THROUGH JUNE 1973

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

JULY 1973

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Waste Disposal
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April through June 1973

Compiled by
A. M. Platt
Nuclear Waste Technology Department

July 1973

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

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INTRODUCTION

This progress report is the third in a series that presents research and development activities in the field of radioactive waste fixation. Previous progress reports were BNWL-1699 and 1741.

SUMMARY

A high-temperature (1400 °C) melt for incorporating PW-6 waste calcine was developed which meets our initial requirements for homogeneity, viscosity and glass leachability. The addition of zinc to the high silica glass substantially reduced the melt viscosity and at the same time improved the melt homogeneity. The glass leach rate appears to be slightly better than that of Pyrex glass.

In fluidized bed calcination studies at ICPP, PW-6 high sodium waste was successfully calcined using a blend of PW-6 waste and stoichiometric quantities of aluminum nitrate. Runs based on substituting $\text{Ca}(\text{NO}_3)_2$ for $\text{Al}(\text{NO}_3)_3$ to form stable sodium compounds were unsuccessful.

A spray calciner and continuous melter assembly has been built and will be used for nonradioactive development of a system for solidifying commercial-type high-level wastes.

The leach rates of Rising Level Glass borosilicate glass core-drilled samples from WSEP PC-9 canister were 1 to 5×10^{-4} fraction of ^{137}Cs leached $(\text{cm}^2/\text{g})^{-1}\text{day}^{-1}$. This canister had been stored almost 2 years at a wall temperature above 390°C , and self-irradiated to a total dose in excess of 10^{11} R, yet the leach resistance of the product had not deteriorated significantly.

A miniature eddy-current probe, which can be lowered into the WSEP canister thermowells, has been constructed and is being tested as a means of pinpointing areas of corrosion in the thermowells.

Initial safety analysis evaluation efforts are associated with the evaluation of a reference system for the management of the high-level wastes arising from the chemical reprocessing of "spent" fuel elements. Currently, the reference waste management system, the modes for failure of the confinement barriers of this reference system, and the potential driving forces for release of radioactivity from the failed system are being defined.

COMMERCIAL WASTE SOLIDIFICATIONLABORATORY STUDIESHigh-Temperature Melt Formulations - J. E. Mendel and
I. M. Warner

Several high-temperature melt formulations were investigated for solidification of PW-6 waste calcine. High-temperature melts will allow higher silica concentrations which are expected to produce more stable and durable glasses. Melts were prepared using the following guidelines. In addition to passing normal tests of homogeneity and leachability, the high-temperature melt should (1) have a viscosity of less than 500 poises at 1400 °C and (2) contain 50 to 60 wt% SiO₂ and 0 wt% B₂O₃.

After testing several variations in composition, a melt was developed which passed our initial requirements including those of homogeneity and leachability. The composition of this melt (No. 73-109) and nine other representative melts investigated are listed in Table 1. As indicated in Table 1, 73-109 was more durable than Pyrex glass in the standard Soxhlet extractor leach test.

Extreme difficulty in attaining the desired viscosity was encountered with the majority of the high-temperature melt formulations investigated. With the addition of zinc to the high silica systems, it was found that not only was viscosity controlled in the desired range but homogeneity of the melt was greatly improved. Apparently, zinc silicate systems facilitate the dissolution of waste calcine in high silica systems.

Further high-temperature studies and investigation of Melt 73-109 will be continued.

TABLE 1. High Temperature Melt Formulations

Sample Number	Composition, wt%								Viscosity at 1400 °C	Leachability % of wt loss at 99 °C			
	SiO ₂	Al ₂ O ₃	CuO	Miscellaneous	Na ₂ O	Fe ₂ O ₃	FP Oxides	Others		1st 24 hr	2nd 24 hr	3rd 24 hr	Total
73-52	59.4	2.3	2.4	0	9.7	8.3	13.2	4.7	Homogeneous	0.57	0.08	0.19	0.84
73-58	59.2	2.5	2.6	3P ₂ O ₅	5.6	1.0	21.1	5.0	Homogeneous	0.24	0.10	0.12	0.45
73-90	60	2.5	2.5	2.5ZrO ₂	9.4	6.0	13.5	3.6	Homogeneous >500 poises	0.41	0.26	0.33	1.00
73-97	51.1	0	0	6.4K ₂ O 12.5ZnO	8.6	5.6	12.5	3.3	Homogeneous <<500 poises	0.30	0.13	0.25	0.69
73-102	57.5	0	0	12.5ZnO	8.6	5.6	12.5	3.3	Homogeneous <500 poises	0.36	0.21	0.15	0.73
73-103	51.1	0	0	12.5ZnO 6.4TiO ₂	8.6	5.6	12.5	3.3	Homogeneous <500 poises	0.40	0.45	0.46	1.31
73-109	51.1	0	0	12.5ZnO 6.4ZrO ₂	8.6	5.6	12.5	3.3	Homogeneous <500 poises	0.22	0.10	0.08	0.40
73-110	52.2	5.0	0	12.8ZnO	8.6	5.6	12.5	3.3	Homogeneous ~500 poises	0.31	0.09	0.15	0.55
73-111	52.2	2.5	0	12.8ZnO	8.6	5.6	12.5	3.3	Homogeneous <500 poises	0.41	0.40	0.18	1.08
73-112	52.2	2.0	0	12.8ZnO 2.5ZrO ₂ 0.5 TiO ₂	8.6	5.6	12.5	3.3	Homogeneous <500 poises	0.54	0.26	0.11	0.90
Pyrex										0.21	0.16	0.18	0.55

Temperature Effects on Leaching Rate - J. E. Mendel and**I. M. Warner**

Temperature effects on leach rates of Glass 72-68 and bottle glass have been investigated. Bottle glass is a glass of commercial composition (see Table 2) and 72-68 is a solidified waste glass prepared with 75% frit 73-1 (see last quarterly report*) and 25 wt% PW-4b calcine. Data were obtained on both glasses by operation of four Paige leaching units at temperatures of 25, 45, 68 and 85 °C. Leach rates are reported in units specified in the last quarterly report,* i.e., fraction of A leached (cm²/g)⁻¹ day⁻¹ where A is the specific ion analyzed. Results of this investigation are shown in Figure 1, where leach rates determined by weight loss and by analysis of the sodium concentration in the leachate are compared.

TABLE 2. Bottle Glass Composition

wt%					
SiO ₂	Al ₂ O ₃	CaO	Na ₂ O	MgO	Fe ₂ O ₃
71	2	10	14	3	Trace

* Quarterly Progress Report, Research and Development Activities, Waste Fixation Program, BNWL-1741, compiled by A. M. Platt, Battelle-Northwest, 1973.

FIGURE 1. Effect of Temperature on Leaching Rate of Bottle Glass and 72-68

In general, bottle glass followed expected behavior with a smooth continuous curve and accelerated leach rates at higher temperatures. The only unusual behavior observed for bottle glass is that the sodium leach rates are lower than the bulk sample leach rates in some instances. Since sodium is generally preferentially leached from glass, it should therefore show higher leach rates than the total sample. Slightly higher leach rates for sodium were observed at temperatures greater than 68 °C.

The leach rate behavior of 72-68 was slightly different than that of bottle glass. One noticeable difference was that the leach rate for 72-68 at higher temperature did not increase as rapidly as the leach rates observed for bottle glass. In addition, the sodium leach rate for 72-68 behaved normally with a higher leach rate than that of the bulk sample.

Devitrification Studies - J. E. Mendel and I. M. Warner

An investigation of devitrification products was begun on two waste glasses which have been chosen for further studies. The two glasses are 72-68 (a low-temperature PW-4b glass) and 73-109 (a high-temperature PW-6 glass).

Devitrification tests were conducted by exposure of the glasses to intermediate temperatures (lower than melting point) for an extended period of time. Melt No. 72-68 was exposed at temperatures of 600 to 850 °C for periods of 7 to 10 days while Melt No. 73-109 was run at temperatures of 800, 850 and 900 °C for a 7-day period. With the aid of x-ray diffraction analysis, zinc orthosilicate (Zn_2SiO_4) was identified as the principle crystalline product in both glasses. A quantitative estimate of Zn_2SiO_4 content was obtained by a dilution technique (see Klug and Alexander, "X-Ray Diffraction Techniques," for details). Results of these analyses are shown in Table 3.

TABLE 3. Zinc Orthosilicate Content of Glasses After Devitrification (wt% of total sample)

	600	650	700	750	800	850	900	% Zn_2SiO_4 Theoretically Possible
72-68	0	0	-	10.4	8.8	6.0	-	31.1
73-109	-	-	-	-	3.7	3.1	7.0	18.3

In addition to Zn_2SiO_4 , 73-109 contained a spinel-like structure (Fe_3O_4 type) which is present in small amounts. The diffraction spectrum of both melts contained additional lines indicating small amounts of other unidentified compounds.

From the data, it appears that maximum devitrification of 72-68 occurs at approximately 750 °C. It also appears that the temperature of maximum devitrification was not reached for 73-109. Further devitrification studies will be conducted to obtain this maximum devitrification temperature.

Ceramic Melter Materials Compatibility with Waste - J. L. Bates and J. J. Rasmussen

Compatibility testing of ceramic refractories in a high temperature waste glass composition have been initiated to select a material for constructing a ceramic melter. A waste glass containing 51.1 wt% SiO_2 , 12.5 wt% ZnO , 6.3 wt% Zr_2O and 30 wt% high-level calcine was made from the virgin oxides. The mixture was held at 1060 °C for 2 hours in a platinum crucible to remove the residual nitrates remaining in the calcine.

The glass was heated in air in a platinum crucible to 1450 °C with the sample suspended in the hot zone above the glass. On reaching the desired temperature, the sample was lowered into the glass; one half of the sample was immersed into the liquid and one half remained out of the liquid. After heating for 48 hours, the sample was removed from the glass and withdrawn slowly from the hot zone of the furnace.

Preliminary results for two samples of fused refractories have been obtained. A sample containing high chromium oxide showed no evidence of corrosion as viewed at low power magnification. Some strips of a brown glassy coating were observed above the glass level on the surface of the sample; however, there was no evidence of attack even at the liquid-gas interface. A high alumina sample (Unicor I*) showed evidence of reaction and dissolution on the portion of the rod in contact with molten glass. Attack was enhanced at the liquid-gas interface.

Testing of other oxide refractories under similar conditions are continuing.

ENGINEERING STUDIES

Developmental Spray Calciner - W. F. Bonner

A spray calciner and continuous melter assembly for engineering-scale solidification of nonradioactive simulated waste has been built in the Engineering Development Laboratory of the 324 Building. Construction is complete, design verification tests have been conducted and operation has begun.

In the spray solidification process liquid waste is continuously converted to a powder in the calciner. The powder is mixed with glass-forming chemicals and melted, then is discharged and cooled in a melt receiving can. The liquid waste is fed through a pneumatic spraying nozzle into the top of a 13-inch-diameter by 6.5-foot tall calciner heated by a 700 °C furnace. The atomized waste is dried and partially calcined before it falls as a powder into the melter below. Calcine which clings to the calciner walls is removed by periodic action of an air vibrator mounted on the calciner. Much of the

* Brand name for sample supplied by Corhart Refractories.

fine powder is carried by the process gases to the adjacent filter chamber where it is retained on a bank of 65 micron pore size sintered stainless steel filters. Accumulated powder caked on the filters is sequentially removed by a blast of heated high-pressure air directed backwards through the filters. The dislodged powder falls into the melter along with the main powder stream.

Frit, continuously added to the melter through a sliding piston solids feeder, mixes with the calcine during melting. Calcine and frit are melted at temperatures up to 1200 °C in an Inconel-690 crucible with 1/2-inch-thick walls prior to being batch discharged through a freeze valve into the melt receiving can. The molten waste is finally cooled at a controlled rate to achieve the desired physical properties of the solid.

Fluidized Bed Calcination Studies - L. L. Taylor (Allied Chemical Corporation). This work was carried out by Allied Chemical Corporation, Idaho Chemical Programs, in support of the Waste Fixation Program.

Ten pounds of simulated PW-4b calcined solids were produced in the 4-inch fluidized bed calciner at the Idaho Chemical Processing Plant and shipped to Pacific Northwest Laboratory (PNL) for glass preparation studies. PW-6 wastes were successfully calcined using a blend of PW-6 wastes and stoichiometric quantities of aluminum nitrate. Runs based on substituting $\text{Ca}(\text{NO}_3)_2$ for $\text{Al}(\text{NO}_3)_3$ to form stable sodium compounds were unsuccessful. The design criteria document for a fluidized-bed rack for use in radioactive demonstrations at PNL is in the final stages of preparation.

PW-4b Wastes

A 75-hour run was completed using a simulated PW-4b waste feed 40% concentrated. The primary objectives of the run were to demonstrate long-term operation and to produce 10 pounds of

'clean' calcine (95% bed replacement) for product glass forming studies being made by Battelle-Northwest. Significant operating conditions and run results are shown in Table 4.

TABLE 4. Operating Conditions and Run Results for PW-4b-4

Feed	PW-4b (40% conc.)
Startup bed	PW-4b-3 (50 wt% PW-4b solids)
Fluidizing velocity, ft/sec	
Above nozzles	1.2
Below nozzles	0.7
Bed Temperature, °C	500 ± 10 °C
Feed rate, l/hr	2-2.5
Waste Nozzle Air Ratio	470
Product bulk density, g/cm ³	2.0
Mean particle size, mm	0.3
Product to fines ratio	2-4
Product agglomeration	Minimal
Attrition resistance index	63
Run length, hr	75
Reason for shutdown	Excessive buildup of undissolved solids in feed system piping.

Approximately 180 liters of feed were calcined during the run with a minimum of difficulty. The run was terminated due to the excessive buildup of undissolved solids in the feed system. Post-run examination revealed a small quantity of feed nozzle agglomerates (normally encountered in 4-inch calciners) on the distributor plate. No operational problems were encountered during the run because of these agglomerates.

PW-6 Wastes

Run PW-6-5. The sodium aluminate flowsheet used in PW-6-1 (stoichiometric quantities of aluminum nitrate added to form NaAlO_2) was calcined at a calcination temperature of 650 °C. Boric acid, which enhances NaNO_3 decomposition in some cases, was present at a concentration of 0.1 M.

Calciner operation was satisfactory, and no severe bed agglomeration was observed; small feed nozzle agglomerates were found on the distributor plate. Operating conditions and significant run results are shown in Table 5.

TABLE 5. Operating Conditions and Run Results for PW-6-5

Feed	PW-6 with AlNO_3
Startup bed	PW-6-1
Fluidizing velocity, ft/sec	
Above nozzles	1.6
Below nozzles	0.9
Bed temperature, °C	650 ± 10 °C
Feed rate, l/hr	1.5
Waste Nozzle Air Ratio	900
Product to fines ratio	8.0
Attrition resistance index	59
Product agglomeration	Minimal
Reason for shutdown	Planned
Run length, hr	12

Run PW-6-6. A 12 hour run was made in which $\text{Ca}(\text{NO}_3)_2$ and $\text{Al}(\text{NO}_3)_3$ were blended with PW-6 waste. Ten liters of 2.1 M $\text{Al}(\text{NO}_3)_3$, 3,225 grams of $\text{Ca}(\text{NO}_3)_2 \times \text{H}_2\text{O}$, and 100 grams of H_3BO_3 were added to each 5 liters of feed and calcined at 600 °C. Post-run examination revealed no agglomerates in the bed, and calciner operation was excellent.

Run PW-6-7. Based on the success of Run PW-6-6, a scoping run was conducted using a flowsheet in which one half of the stoichiometric amount of $\text{Al}(\text{NO}_3)_3$ was replaced with $\text{Ca}(\text{NO}_3)_2$ and calcined at 600 °C. The run was terminated due to severe agglomeration in the bed.

AEC WASTE SOLIDIFICATIONSOLIDIFICATION EQUIPMENT STUDIES - J. D. Kaser

In order to choose appropriate process equipment for solidifying In-Tank-Solidification (ITS) waste, the ultimate scale of operation must be taken into consideration. It has been estimated that the final throughput capacity will be about 3.7 million gallons per year of ITS salt cake which is equivalent to a vitrified product flow of about 21,000 pounds per hour, or 4.5×10^4 tons/yr over a 10-year period, which is orders of magnitude greater than that required for commercial power reactor waste solidification equipment.

In order to gather additional ideas on large-scale processing equipment, several industrial facilities in the Northwestern United States were visited to obtain technical information on large-scale production of glass and bricks. These facilities were: Penberthy Electromelt and Northwest Glass Works of Seattle, Washington; Columbia Brick Works, Portland, Oregon; and Albany Metallurgy Research Center, Albany, Oregon. Of these, Penberthy is most interested in the AEC waste problem since they are in the business of making melters and consulting with the glass industry.

In the Penberthy electric resistance melting process, electrodes are immersed in the melt and heat is generated by the electric current passing through the melt. The electrodes are made of molybdenum unless the melt is oxidizing, then the electrodes are made of tin oxide. The melter is started up with a gas or oil burner. This type of melter can be scaled up to very large capacities by simply increasing the number of electrode pairs. Scaling down is more of a problem. A proper voltage-to-electrode separation ratio must be observed in changing size. The electrodes are subject to wear and their spacing

must be periodically adjusted by pushing the electrodes into the crucible after melting the frozen glass seal by turning off the cooling water to the electrodes.

RADIOACTIVE DEMONSTRATIONSSOLIDIFICATION CELL (B-CELL) REACTIVATION - B. O. Kahle

Reactivation of B-Cell in preparation for radioactive demonstrations is continuing with the major overhauling of the in-cell cranes completed. Instrument checkout of the in-cell spray solidifier indicated that essentially all the control thermocouples are operable.

RADIOACTIVITY FOR WFP DEMONSTRATION RUNS - D. H. Siemens

A portion of the radioactivity that is required for the WFP demonstration runs will be obtained from commercial high-level waste. This waste will be received at Pacific Northwest Laboratory (PNL) in the reprocessor's canister as a fluidized bed calcine. The radioactive waste will have been calcined to form a coating on an inert bed material. The canister, containing the calcine, will be moved into the solidification cell (B-Cell). The canister will then be opened and the calcine transferred to a vessel for subsequent dissolution of the radioactive waste with dilute nitric acid. The remaining inert material will be returned to the canister for disposal. Preparations are currently being made for shipping the calcined waste to PNL and for subsequent processing of the calcine in B-Cell.

PRODUCT CHARACTERIZATIONMEASUREMENTS ON CORE-DRILLED SAMPLES - J. E. Mendel

The canister of borosilicate glass prepared in WSEP Run PC-9 was core drilled in three locations after 2.5 years of interim storage (Core No. 67, 13 inches from bottom, Core No. 68, 18 inches from bottom, and Core No. 69, 32 inches from the bottom). This 6-inch-diameter 310 stainless steel canister contains borosilicate glass made by the Rising Level Glass process. The glass contains only about 18 wt% SiO_2 , but contains about 47 wt% fission product oxide. The remainder of the composition is mainly B_2O_3 and Na_2O ; thus the glass would be expected to have a relatively poor chemical durability.

This canister was one of the hottest canisters produced in the WSEP program, initially containing 310 W/liter of fission product heat. Prior to core drilling the canister had been stored in heated air in a SSETF pod for 22 months. The thermal history of the canister is shown in Figure 2. Core samples 67 and 68, from the hottest area of the canister appeared completely crystalline, i.e., devitrified, by visual inspection. Sample 69, from about 3 inches below the top surface of the product, and thus in a somewhat cooler area, retained some glassy characteristics.

PC-9 Leak Check. The PC-9 canister could not be leak checked since one of the thermowells had been penetrated during previous core drilling in December 1970.

PC-9 Thermal Conductivity. Even though almost completely devitrified the thermal conductivity of the solidified waste in PC-9 canister has not changed significantly from the initial value of about $1.0 \text{ W}(\text{cm}^2)^\circ\text{C}/\text{m}$. Figure 2 shows the change in Δt , centerline to wall, for the PC-9 canister during the time it

FIGURE 2. Thermal History of PC-9 Canister

has been in interim storage. The rate of decrease in Δt corresponds closely to that which would be predicted from the decay rate of ^{144}Ce , the major heat-producing radioisotope in the canister.

PC-9 Leach Rate. Some preliminary leach data for core samples from PC-9 canister are shown in Table 6.

TABLE 6. Leachability of WSEP Rising Level Borosilicate Glass Product After 1.8 Years Storage at 460 to 390 °C Wall Temperature

Core Sample	Radial Location	Leach Rate After 1 Week in Distilled H_2O at 25 °C. Fraction ^{137}Cs Leached
		$(\text{cm}^2/\text{g})^{-1}\text{day}^{-1}$
68A	Wall	1.1×10^{-4}
68C	Centerline	5.1×10^{-4}
69A	Wall	4.3×10^{-4}
69C	Centerline	4.5×10^{-4}

These results are very similar to those obtained previously (see Figure 5.23, BNWL-1666). It may be concluded that the leaching properties of the RLG process borosilicate glass have not deteriorated significantly even after severe temperature treatment (almost 2 years at wall temperatures above 390 °C and centerline temperatures of approximately 500 to 700 °C) and radiation exposure (in excess of 10^{11} R).

PC-9 Wall Thickness Measurements. The wall thickness was measured ultrasonically. No significant changes from the initial wall thickness measurements were found.

THERMOWELL LEAKS IN WSEP CANISTERS - J. E. Mendel

A helium mass spectrometer vacuum leak check is made routinely prior to core drilling the WSEP waste canisters. The leak checks have revealed leaks in the thermowells of 4 of

the 14 WSEP waste canisters core drilled to date, and in one other canister which has not been core drilled. The latest canister to be found leaking is SS-13, which passed the routine after-fill leak test on July 28, 1970, 2.5 months after filling, yet was found to leak at the thermowell, in the precore-drill leach test made June 8, 1973. This behavior is similar to that of the other canisters in which leaks have been found, in that the leak developed during interim storage, and not during filling when peak temperatures were encountered (see Table 7).

TABLE 7. Leak Testing of WSEP Canisters

WSEP Run No.	Process (a)	Canister Description	Date Canister Filled	Date and Result of First Leak Check (b)	Date and Result of Second Leak Check (c)	Interim Storage Conditions
PG-6	Phosphate Glass	8-inch diam 304L SS	May 15, 1968	Sept. 10, 1969 1.5×10^{-7}	Jan. 14, 1971 Both thermowells leaked	Water at $\sim 50^\circ\text{C}$
PG-7	Phosphate Glass	8-inch diam 310 SS	July 10, 1969	July 23, 1969 4.1×10^{-7}	July 18, 1970 Leak at thermowell	Air at $\sim 400^\circ\text{C}$
SS-5	Spray Solidification--Phosphate ceramic	8-inch diam 304L SS	Oct. 21, 1968	May 7, 1970 Leak at thermowell	---	Water at $\sim 50^\circ\text{C}$
SS-12	In-pot Melting Borosilicate Glass	8-inch diam 304L SS	April 7, 1970	May 8, 1970 8.0×10^{-7}	Dec. 1, 1970 Both thermowells leaked	Air at $\sim 50^\circ\text{C}$
SS-13	In-pot Melting Borosilicate Glass	8-inch diam 304L SS	May 12, 1970	July 28, 1970 4.1×10^{-7}	June 8, 1973 Both thermowells leaked	Water at $\sim 50^\circ\text{C}$

a. Pot calcination process canisters are not leak checked because of outgassing problem with calcine product.

b. Vacuum helium mass spectrometer leak rate, atm cm^3/sec .

c. Thermowells are 1/2 or 3/4-inch Schedule 80 310 stainless steel pipe in all cases.

It should be emphasized that the leaks which have been found in the WSEP canisters are all in the thermowells, and not in the canister walls. Ultrasonic wall thickness scanning, and examination of the circular wall segments removed during core drilling, indicate there has been no significant corrosion of the canister walls; thus, if the thermowells were plugged, or totally absent, as will probably be the case in commercial waste canisters, canister integrity would remain. It is important, however, to understand the reasons for the thermowell failures in order to (1) remedy the conditions which caused the

failures in any future waste canisters equipped with thermowells, and (2) make certain that the failure mechanisms pertain only to the thermowells and cannot under certain circumstances also cause leaks in the canister walls.

The information obtained to date indicates that wide-spread corrosion of the thermowells has not occurred. Instead the leaks are probably due to cracks or isolated areas of accelerated corrosion. Several samples of thermowells have been obtained by core drilling and none have shown significant corrosion. A small eddy-current probe has been designed which can be lowered into the thermowells. It will be used to pinpoint areas of likely corrosion for future core sampling of the thermowells.

SAFETY ANALYSIS

W. K. Winegardner

The purpose of the safety analysis task is to define and investigate waste management systems in relation to the containment, immobilization, and isolation of hazardous wastes and to evaluate the radiological safety implications of these systems. Early efforts have been primarily related to the definition of a reference, state-of-art waste management system, the modes for failure of the confinement barriers of this reference system, and the potential driving forces for the release of radioactivity from the failed system.

REFERENCE SYSTEM DESCRIPTION

Details of the reference waste management system selected for initial safety analysis evaluations are summarized in Figure 3 and Table 8. Retrievable 5-year storage of high-level liquid wastes will be followed by solidification and encapsulation and an additional 5 years of retrievable storage of the resulting canisters at the fuel reprocessing plant. Wastes will then be shipped "offsite" to an interim (100 year) storage facility, again for storage in a retrievable manner. The safety of the candidate ultimate disposal concepts, which must follow the 100-year interim storage are under investigation in a similar evaluation program.

The waste characteristics shown in Table 8 are based primarily on information from Nichols et al.* for the aggregate of wastes accumulated through the year 2000; the table shows the assumed waste distribution within the reference system in the year 2000.

* J. P. Nichols et al., Projections of Fuel Reprocessing Requirements and High Level Solidified Wastes from the U.S. Nuclear Power Industry, ORNL-TM-3965 (draft), to be published.

- (a) ASSUMES 0.5% OF U AND Pu IN LWR AND LMFBR PLANT FUELS, 0.5% OF U AND Th IN HTGR FUEL, AND 100% OF ALL OTHER ACTINIDES IN WASTE.
 (b) STORAGE IN NONBOILING, WATER-FILLED BASINS.
 (c) ASSUMES 2 FT³/MT, 6 FT³/MT, AND 3 FT³/MT SOLIDS FOR LWR, HTGR, AND LMFBR PLANT FUELS, RESPECTIVELY.
 (d) ASSUMES CANISTERS, 12 INCHES DIAMETER x 10 FEET LONG, EACH CONTAINING 6.28 FT³ OF SOLIDS.

FIGURE 3. Reference Waste Management System

TABLE 8. (a) Selected Waste Characteristics--
Reference Waste Management System

	Reprocessing Load (MT of fuel) (b)				Radioactivity (Ci) (c)		Thermal Power (Watts) (c)		Volume (ft ³) (b)		No. of Canisters (b)
	Total	LWR	HTGR	LMFBR	Fission Products	Actinides	Fission Products	Actinides	Liquid	Solid	
Liquid Storage (at each Fuel Reprocessing Plant)	9,650	4,225	1,787	3,638	8.6×10^9	6.6×10^7	3.2×10^7	1.6×10^6	4.6×10^5	-	-
Solid Storage (at each Fuel Reprocessing Plant)	7,231	5,249	1,088	894	5.0×10^9	1.7×10^7	2.0×10^7	2.0×10^5	-	2.0×10^4	3,139
Transportation (one cask load)	28.3	23.3	-	-	8.8×10^6	6.6×10^4	2.9×10^4	2.0×10^3	-	5.7×10^1	9
Solid Storage (AEC Federal Repository)	68,581	64,911	3,129	541	5.3×10^{10}	1.2×10^9	2.3×10^8	4.1×10^7	-	1.50×10^5	23,917

a. See Figure 3 for assumptions used in the development of this table.

b. Based on "actual" reprocessing load (from 181,000 MT of accumulated spent fuel, including low exposure start-up fuel, scrap, etc.) from J. P. Nichols et al., Projections of Fuel Reprocessing Requirements and High-Level Solidified Wastes from the U.S. Nuclear Power Industry, ORNL-TM-3965 (draft), to be published.

c. Based on equivalent reprocessing load of 167,000 MT of accumulated spent fuel at rated exposures.

Confinement barriers associated with various operations of of the reference waste management system are summarized in Figure 4. As shown in this figure, at least two engineered barriers will be provided to prevent the release of liquid wastes. Not shown is the fact that surrounding soil can also act as a confinement barrier simply as the result of the soil's sorption properties. Solidification processing will be conducted in shielded hot cells that have thick concrete walls. Solid (canister) storage at both the reprocessing plant and the interim storage facility will be based on nonboiling, water-filled basins. These solid storage basins are assumed to be modular concrete structures, located below grade. Finally, except for shipping, it is expected that final ventilation confinement of radioactivity will be provided by filtration.

CONFINEMENT BARRIER FAILURE MODES

A variety of events can be pictured that could conceivably void the confinement or containment afforded by the barriers shown in Figure 4. Potential failure modes are categorized as either geologic events, meteorological-hydrological events, or uncontrolled processes. The latter includes chemical, thermal,

BARRIERS TO
AIRBORNE RELEASEBARRIERS TO LIQUID
AND/OR SOLID RELEASE

- (a) OTHER EXHAUST GAS TREATMENT WILL PROBABLY BE INSTALLED UPSTREAM OF FILTERS.
 (b) POSSIBLY NOT PROVIDED OR NOT A CONTAINMENT/CONFINEMENT BARRIER IN EVERY INSTANCE.

FIGURE 4. Reference Waste Management System
 Confinement/Containment Barriers

mechanical, and nuclear phenomena. Many of these uncontrolled processes or their "initiating" event can be directly or indirectly related to man's activities.

Geologic Events

Only short-term or catastrophic geologic events will be considered here simply because of the interim nature of the facility and the relatively short lifetime of the engineered barriers, i.e., 20 to 100 years. The relationship between a particular storage site and long-term geologic processes such as glacial erosion and denudation would, of course, become important if it is possible for barrier failure to result in waste deposition into the environment in an irretrievable manner.

- Earthquakes

Consideration of potential short-term geologic driving forces (failure mechanisms) immediately brings to mind the sudden movement of layers of the earth's crust by seismic activity. Obviously, the cracking, warping, and tilting associated with earthquakes could negate containment barriers of storage and solidification facilities if the barriers were improperly designed. Protection against this failure mechanism is obtained by proper siting and facility design.

- Volcanic Activity

Volcanic activity is the second short-term geologic process that should be considered as a potential failure mode. Like seismic activity, volcanic action is determined by tectonic conditions. Protection against this failure mechanism will probably be obtained solely by siting.

Meteorological-Hydrological Events

As in the case of geology, consideration is limited to short-term, violent severe meteorological-hydrological events rather than long-term phenomena, such as climatic changes. Specifically, wind and flooding are envisioned as the primary driving forces for release of radioactivity.

- Wind

Wind storms, tornadoes, and hurricanes all could destroy the integrity of certain of the engineered confinement barriers. In addition, tornado-driven missiles and the possibility of hydrostatic water removal by the tornado event are conceivable failure mechanisms.

- Floods

Floods could conceivably negate confinement barriers, especially those associated with either liquid storage or ventilation exhaust systems. Again, siting of permanent facilities is the major protection; weather history and site selection in relation to major surface water supplies are the primary considerations. The latter, consideration of surface supplies, would include not only natural sources but the evaluation of man-made reservoirs and the accompanying possibility of dam failure.

Uncontrolled Processes

Table 9 lists several chemical, thermal, mechanical, and nuclear processes that, when uncontrolled, could lead to loss of confinement. "Initiating" events for the various reactions are also postulated. Obviously other contributing factors could accompany and even precede the so-called "initiating" events. As suggested earlier, many of these "initiating" events can be related to man's activities.

TABLE 9. Uncontrolled Processes that Could Lead to Failure and/or Penetration of Confinement Barriers

<u>Process</u>	<u>Postulated "Initiating" Events</u>
Fire and/or Explosion	Transportation accident Airplane crash Sabotage Formation of radiolytic hydrogen Organic contamination of liquid wastes Nuclear weapon Reestablishing cooling after loss-of-cooling incident
Impact	Failure of canister handling system, dropping canister Transportation accident Massive missile, e.g., airplane crash
Corrosion	Local corrosion of liquid storage tanks Contamination of canister coolant Changes in microstructure of canister material as a result of long exposure to temperature and radiation
Pressurization	Gaseous radiolytic decay products in canister
Fusion	Canister penetration as a result of prolonged loss of cooling
Criticality	---

DRIVING FORCES FOR RELEASE OF RADIOACTIVITY

The following well-known and frequently cited phenomena are the basic mechanisms for the release of radioactivity in a form that can be transported to man's environment.

- Dissolution (leaching)

Canister failure during basin storage, e.g., by corrosion, could result in water-solid waste contact and the accompanying possibility of leaching radioactive

constituents from the solid. The resulting contamination can, in many instances, be removed from the water, e.g., by ion exchange. In addition, transportation accidents can be postulated where failed canisters are exposed to surface water supplies.

- Fluid Flow

Failure of all liquid storage system barriers would result in the release of radioactivity by fluid flow. The extent of the release, however, will be governed not only by the degree of failure and the filtering action of surrounding soil but also by the solubility of the solid waste and the degree of solid-liquid phase interactions as the solute moves through the upper layer(s) of the earth. Liquid release can be described as a transport as well as a release mechanism.

- Temperature Driven Release

High temperatures could cause failure of the confinement barrier and/or could volatilize radioactivity from the solids as well as the liquid high-level wastes. Transportation accidents that involve a high-temperature fire and where canister integrity is lost can be postulated. Cooling systems are associated with various operations of the reference system and prolonged cooling system malfunction could conceivably initiate significant temperature transients, especially during early years of storage.

- Dispersion by Impact or Explosion

Finally, it is conceivable that the granular or monolithic solid wastes could be dispersed by impact or explosion. Obviously, the monolithic solid would probably be fragmented into small, readily dispersible particles by the event.

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