

ENGINEERING-SCALE VITRIFICATION OF COMMERCIAL HIGH-LEVEL WASTE

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INTRODUCTION

The acceptance of nuclear power is currently being restrained by the need for acceptable technology to immobilize and dispose of nuclear wastes. To provide this technology, major programs have been conducted at the Pacific Northwest Laboratory (PNL), which is operated by Battelle Memorial Institute for the Department of Energy. Although detailed program objectives have changed over the years, the continual goal at PNL has been the development of waste immobilization technology. To date, technology for immobilizing commercial high-level waste (HLW) has been extensively developed, and two major demonstration projects have been completed, the Waste Solidification Engineering Prototypes (WSEP) Program and the Nuclear Waste Vitrification Project (NWVP).

The feasibility of radioactive waste solidification was demonstrated in the WSEP program between 1966 and 1970 ¹ using simulated power-reactor waste composed of nonradioactive chemicals and HLW from spent, Hanford reactor fuel. Thirty-three engineering-scale canisters of solidified HLW were produced during the operations. In early 1979, the NWVP demonstrated the vitrification of HLW from the processing of actual commercial nuclear fuel. This program consisted of two parts, 1) waste preparation and 2) vitrification by spray calcination and in-can melting. This report presents results from the NWVP.

WASTE PREPARATION

A pilot-scale hot cell facility was assembled to obtain HLW from spent, commercial nuclear reactor fuel. A conventional

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purex-type process was used to assure generation of waste that was representative of potential commercial HLW.

Spent Fuel

Spent light-water reactor (LWR) fuel from which the HLW was generated came from the storage pool at Nuclear Fuel Services' West Valley, New York, plant. Six zircalloy-clad, 14 x 14 style bundles that had been irradiated at the Point Beach Reactor facility were used. General data on the spent fuel is given in Table I.

TABLE I. Spent Fuel Average Data

Parameter	Bundles 1 to 3	Bundles 4 to 6
Reactor	Point Beach	Point Beach
Fuel type	LWR	LWR
Configuration	14 x 14 (179 fuel rods)	14 x 14 (179 fuel rods)
Initial enrichment	2.27 wt% ²³⁵ U	3.04 wt% ²³⁵ U
Final enrichment	0.85 wt% ²³⁵ U	0.94 wt% ²³⁵ U
Discharge date	Oct. 10, 1972	Apr. 10, 1974
Burnup	20,500 Mwd/tU	29,228 Mwd/tU
Average power	29 MW	30 MW
Waste extraction	Dec. 1978 to Jan. 1979	Feb. to Mar. 1979

The six spent fuel assemblies were transported to PNL using a Nuclear Assurance Corporation style shipping cask. All six bundles were received and placed in a storage rack inside a hot cell before any shearing or dissolution was begun. The PNL facility for receiving, shearing and dissolving the fuel is shown in Fig. 1. Fuel receiving and shearing for NWVP is discussed by Siemens ².

Fuel Disassembly and Dissolution

As feed for the dissolver was needed, a fuel assembly was placed on a disassembly table. The top nozzle plate was removed first by severing, with an inside tubing cutter, each of the 17 control rods and instrument tubes. The 179 zircalloy fuel rods that are not attached to the fuel bundle assembly heads at either end were then pulled from the assembly and fed into a multiple tube puller with a master-slave manipulator. The fuel

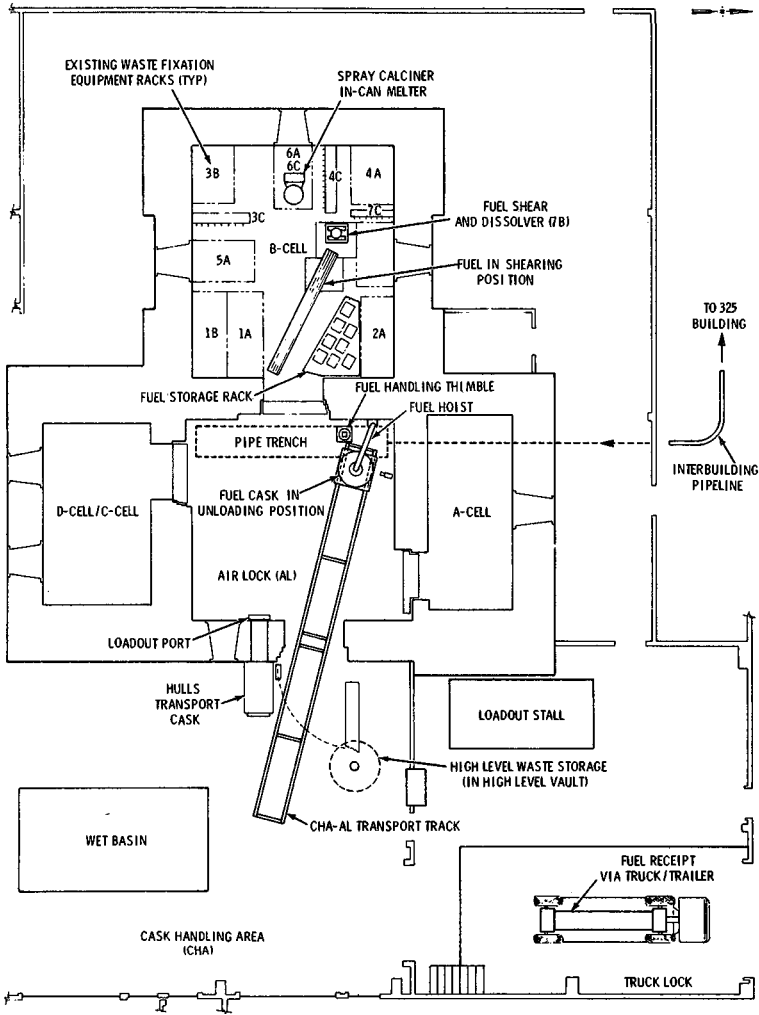


FIG. 1. PNL Radiochemical Engineering Cells (324 Building)

rods were fed into a hydraulic shear. Sheared pieces fell into the fuel dissolver. The three low-burnup fuel assemblies were disassembled and sheared first.

During a normal batch operation, 40 to 42 rods representing 86.4 kg of U were sheared and dissolved. Twenty-six separate dissolver batches were required to process the six LWR fuel assemblies. The dissolver solution was transferred to a holding tank through a 100- μ -pore, sintered stainless steel filter. Two filters were used for the 26 batches to collect dissolver solution solids for possible future characterization. The leached, spent fuel hulls were dried in a furnace and were then packaged in 5-gal., carbon steel, crimp-top buckets. The buckets were placed in a shielded shipping cask and were shipped to retrievable storage operated by Rockwell Hanford Company.

Waste Extraction

A Purex-type solvent extraction system for the first co-decontamination cycle was selected for NWVP. In this system, the aqueous waste stream from the extraction column contains essentially all the fission products, solids and other non-extractable impurities present in the dissolver solution. This stream becomes the HLW from the reprocessing system. For NWVP, the solvent extraction system consisted of an extraction column along with a strip column and two organic wash columns. The dissolver solution was fed to the solvent extraction system based on a flowsheet rate of 50 kg U/d.

The equipment in the solvent extraction system operated quite well with only one major problem during the three months of operation. During the first three weeks of operation, including the hot startup, the equipment operated with no apparent difficulty. Thereafter, an overflow line intermittently plugged, which restricted the flow rate and required cleanout. Time spent on this problem delayed completion of the project by three to four weeks and reduced on-line efficiency from 90% before the plugging started to 62% for the overall project. Since that was the only significant problem encountered in the operation of the new facility, the overall performance was considered quite satisfactory.

The waste stream from the solvent extraction system was prepared for the ion-exchange partitioning system by concentrating the solution to 7.5M HNO₃. The anion ion-exchange system consisted of two separate systems; the first system was installed and operated in the hot cell, while the secondary system was placed in a contact-operated, shielded glovebox. Each system had a primary column sized to contain 450 g Pu/batch. Each system

also had a second column as a backup or tailing column. The fission product decontamination factor for the first ion-exchange system was high enough that the plutonium product could be calcined with no further purification. The uranium-bearing streams from the ion-exchange system, along with the streams from the off-gas scrubber, the solvent wash system and the calciner off-gas condenser, were collected and sampled. These solutions were then transferred via a shipping cask by the Rockwell Hanford Operations for disposal.

The plutonium nitrate product from the ion-exchange system was directly calcined to an oxide in a continuous-fed screw calciner. This work is discussed by Bryan³. The plutonium oxide product from the calciner was cooled, weighed and then stored. Batches of approximately 2.5 kg were homogenized in a V-blender, sampled, reweighed and then stored.

High-Level Liquid Waste

Different wastes were used for each of the two NWVP vitrification runs. The low burnup fuel was used for Run 1. Run 2 consumed waste from the high burnup fuel and also utilized tank heels from Run 1. The reprocessing of spent fuel equivalent to 1024 kg of original U for the first NWVP vitrification run produced 8370 L of HLW, or about 8175 L/tU. Since it was desired to feed the spray calciner with HLW at 850 L/tU, a nearly 10-to-1 concentration was necessary. The processing scheme for concentrating the HLW is described by Wheelwright⁴. The second NWVP vitrification run consumed waste from processing spent fuel representing 860 kg of U in the original fuel. Dilute HLW (5130 L) was concentrated to 650 L for calcination.

During concentration, acid stripping was necessary to prevent potential ruthenium volatility at high HNO_3 concentrations. The nominal dilute waste was batch-concentrated from 1.5M to 4M HNO_3 . Acid stripping was accomplished by adding strip water to control acid concentrations during continual concentration.

The calculated HLW fission product and actinide concentrations are shown in Tables II and III. Inert, nonradioactive chemicals and uranium were added to the radioactive HLW to simulate the presence of intermediate-level wastes, corrosion products and degraded solvent. Inert additives, the same for both runs, are listed in Table IV.

TABLE II. Calculated Waste Constituents for Run 1 (Basis 1 tU)

Element	Grams	Moles	Oxide Formed	g Oxide
<u>Fission Products</u>				
Ge	0.22	0.003	GeO ₂	0.3
As	0.05	0.0003	As ₂ O ₃	0.07
Se	31.7	0.40	SeO ₂	44.5
Rb	203.0	1.19	Rb ₂ O	222.0
Sr	503.0	5.74	SrO	594.8
Y	284.0	1.60	Y ₂ O ₃	360.7
Zr	2,280.0	24.99	ZrO ₂	3,079.8
Mo	2,130.0	22.20	MoO ₃	3,195.9
Tc	530.0	2.68	Tc ₂ O ₇	829.8
Ru	1,350.0	13.36	RuO ₂	1,777.8
Rh	288.0	1.40	Rh ₂ O ₃	355.2
Pd	838.0	7.88	PdO	964.0
Ag	41.7	0.19	Ag ₂ O	44.8
Cd	45.3	0.40	CdO	51.7
In	1.0	0.009	InO	1.1
Sn	31.5	0.27	SnO ₂	40.0
Sb	7.7	0.03	Sb ₂ O ₃	9.2
Te	348.0	2.73	TeO ₂	435.3
Cs	1,530.0	5.76	Cs ₂ O	1,622.0
Ba	975.0	7.10	BaO	1,089.0
La	782.0	2.81	La ₂ O ₃	917.1
Ce	1,520.0	10.85	CeO ₂	1,867.0
Pr	738.0	0.87	Pr ₆ O ₁₁	891.6
Nd	2,520.0	8.74	Nd ₂ O ₃	2,939.0
Pm	22.1	0.08	Pm ₂ O ₃	25.7
Sm	571.0	1.90	Sm ₂ O ₃	662.2
Eu	95.3	0.31	Eu ₂ O ₃	110.4
Gd	54.3	0.17	Gd ₂ O ₃	62.6
Tb	1.2	0.002	Tb ₄ O ₇	1.4
Dy	0.8	0.002	Dy ₂ O ₃	0.9
Subtotal	<u>17.7 × 10³</u>			<u>22.20 × 10³</u>
<u>Actinides</u>				
U ^(a)	4,850.0	6.79	U ₃ O ₈	5,719.0
Np	252.0	1.06	NpO ₂	286.0
Pu	38.4	0.16	PuO ₂	43.5
Am	84.3	0.35	AmO ₂	95.5
Cm	4.6	0.01	Cm ₂ O ₃	5.1
Subtotal	<u>5.23 × 10³</u>			<u>6.15 × 10³</u>
TOTAL	<u>22.95 × 10³</u>			<u>28.35 × 10³</u>

(a) Addition of depleted U required to achieve this value.

TABLE III. Calculated Waste Constituents for Run 2 (Basis 1 tU)

Element	Grams	Moles	Oxide Formed	g Oxide
<u>Fission Products</u>				
Ge	0.33	0.005	GeO ₂	0.5
As	0.07	0.0005	As ₂ O ₃	0.1
Se	4.65	0.06	SeO ₂	6.5
Rb	307.0	1.80	Rb ₂ O	335.7
Sr	758.0	8.65	SrO	896.4
Y	422.0	2.37	Y ₂ O ₃	535.9
Zr	3,330.0	36.51	ZrO ₂	4,498.0
Mo	3,100.0	32.31	MoO ₃	4,651.0
Tc	759.0	3.83	Tc ₂ O ₇	1,188.0
Ru	1,920.0	19.0	RuO ₂	2,528.0
Rh	369.0	1.79	Rh ₂ O ₃	455.0
Pd	1,210.0	11.37	PdO	1,392.0
Ag	53.7	0.25	Ag ₂ O	57.7
Cd	69.1	0.61	CdO	78.9
In	1.2	0.01	InO	1.4
Sn	44.8	0.38	SnO ₂	56.9
Sb	11.4	0.05	Sb ₂ O ₃	13.6
Te	502.0	3.93	TeO ₂	627.9
Cs	2,240.0	8.43	Cs ₂ O	2,375.0
Ba	1,420.0	10.34	BaO	1,585.0
La	1,140.0	4.10	La ₂ O ₃	1,337.0
Ce	2,230.0	15.91	CeO ₂	2,739.0
Pr	1,080.0	1.28	Pr ₆ O ₁₁	1,305.0
Nd	3,680.0	12.76	Nd ₂ O ₃	4,293.0
Pm	37.0	0.13	Pm ₂ O ₃	43.0
Sm	803.0	2.67	Sm ₂ O ₃	931.2
Eu	150.0	0.49	Eu ₂ O ₃	173.7
Gd	91.2	0.29	Gd ₂ O ₃	105.1
Tb	1.58	0.002	Tb ₄ O ₇	1.9
Dy	0.92	0.003	Dy ₂ O ₃	1.9
Subtotal	25.7 x 10 ³			32.2 x 10 ³
<u>Actinides</u>				
U ^(a)	4,800.0	6.72	U ₃ O ₈	5,660.0
Np	426.0	1.80	NpO ₂	483.5
Pu	40.6	0.17	PuO ₂	46.0
Am	297.0	1.23	AmO ₂	336.4
Cm	11.4	0.02	Cm ₂ O ₃	12.5
Subtotal	5.58 x 10 ³			6.54 x 10 ³
TOTAL	31.3 x 10 ³			38.7 x 10 ³

(a) Addition of depleted U required to achieve this value.

TABLE IV. Inert Chemicals Added to HLW (kg/tU)

Compound	Run 1	Run 2
NaNO ₃	23.67	34.91
Fe(NO ₃) ₃ 9H ₂ O	84.55	124.71
Cr(NO ₃) ₃ 9H ₂ O	3.72	5.49
Ni(NO ₃) ₂ 6H ₂ O	1.55	1.99
H ₃ PO ₄ (75%)	1.52	2.24

SPRAY CALCINATION/IN-CAN MELTING

Process and Equipment

The PNL in-cell spray calciner and in-can melter are shown schematically in Fig. 2. The spray calciner is a heated, vertically mounted chamber 35.6 cm in dia by 197.5 cm high (14 in. in dia by 77.8 in. high) into which liquid waste is sprayed and dried. The calciner spray chamber walls are heated to about 750°C by a 3-zone, 45-kW resistance furnace surrounding the chamber. Compressed air atomizes the HLW feed solution as it enters the top of the spray chamber through a commercially available spray nozzle.

As atomized waste droplets fall through the chamber, heat is transferred from the walls by radiation and convection. As it drops through the chamber, the waste is concentrated, dried and oxidized to form a fine calcine product of typically less than 0.5% moisture and 1% nitrate. Buildup of solids on the chamber walls is prevented by periodic use of a wall-mounted, air-operated vibrator. Porous, sintered stainless steel filters remove solid particles from gases leaving the spray chamber. The off-gas filter unit contains sixteen 5.1-cm-dia by 91-cm-long (2-in.-dia by 36-in.-long) filters with a nominal pore size of 65 μ. The calciner is operated at a nominal pressure of -12 to -25 cm (-5 to -10 in.) of water.

Glass-forming material called frit is metered into the lower cone of the spray calciner at a rate proportional to the rate of calcine production. Frit falls with the calcine into the canister below the calciner, where the mixture is melted and then cooled to form a borosilicate glass. With in-can melting, vitrification is accomplished using the canister as the melting crucible. The canisters for NWVP were 20 cm in dia by 244 cm high (8 in. in dia by 8 ft tall). Details of the canister are presented by Bjorklund and Hanson⁵. The melter furnace is a

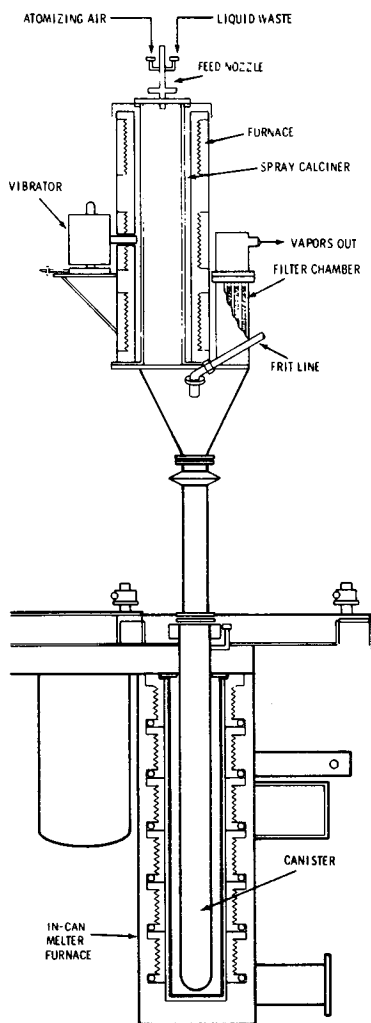


FIG. 2. In-Cell Spray Calciner/In-Can Melter

6-zone, resistance type. Each zone is 35.6 cm (14 in.) high and delivers 15 kW of power. The furnace operates at 1050°C.

Operating Experience

Both vitrification runs proceeded smoothly, including operation of the effluent cleanup system. One significant mishap occurred that affected operations. This mishap can be readily avoided in the future. A weld was mistakenly omitted during the assembly of the calciner nozzle; consequently, the ceramic insert in the nozzle broke loose during Run 1. Due to an error in the feed system design, this failure was not noticed until after the run. Without the insert, little atomization of the liquid feed occurred, which resulted in a buildup of dried waste on the lower cone of the spray calciner. This explained a lower-than-expected glass volume in Canister 1. The material was remotely removed from the calciner cone before Run 2. A small amount of this material plus some frit was added to the second canister before Run 2.

All other processing equipment operated well. The sintered-metal, off-gas filters removed the entrained calcine dust with little further drop in pressure. Filters were blown back every 20 min, and the spray chamber was vibrated every 30 min. Pumps, furnaces, frit feeders and auxiliary equipment operated smoothly during both runs.

The waste-glass canisters were held at operating temperature during the entire filling operation. At the end of each run, the filled canister was cooled in the furnace with forced air until the canister wall temperatures reached 550°C; this process took about 2 h. The canister walls were held at 550°C for 6 h, and the canister was then allowed to cool naturally.

Selected data for the two vitrification runs is given in Table V.

TABLE V. High-Level Waste and Processing Data

Characteristic	Value	
	Run 1	Run 2
Calciner Feed		
Concentration, L/tU	833	760
HNO ₃ , M	4.4	6.6
Solids content, g oxide/L	66	86
Heat content, W/L	1.12	2.05
Activity, Ci/L	313	526
Feed processed, L	806	430
Average feed rate, L/h	15.7	12.7
Run time, h	53	33.6
Average fuel burnup, MWd/tU	20,400	25,900
Onstream factor, %	99	96
Glass-Filled Canister		
Net glass weight, kg	116	145 ^a
Net calcine added, kg	22.3	37.0
Net frit added, kg	93.7	83.7
Estimated bulk density, g/cm ³	2.8	2.7
Fill height, cm	137	175
Glass volume, L	41.3	52.9
Frit-to-Calcine ratio	4.2:1	2.2:1
Glass density, g/cm ³	2.8	2.7
Equivalent uranium fuel, tU	0.40	0.72
Decay heat, ^b W	380	1,010
Activity, ^b Ci	1.05 x 10 ⁵	2.64 x 10 ⁵

a Includes 8.2 kg calcine and 16.3 kg frit added to the canister during spray calciner cleanout.

b Based on ORIGEN computer code predictions.

Melt Level Detection

During NWVP, intrinsic gamma emissions were monitored to determine the melt level in the in-can melter. In this technique, collimators that partially penetrate the cell wall provide view points at various canister heights, as shown in Fig. 3. Gamma radiation intensity at the detector drastically increases as the melt level rises past the collimator view point. Thus, the time at which the melt level reaches the view point is known. As many view points as needed can be used. This technique is applicable to many radioactive processes needing level detection capabilities.

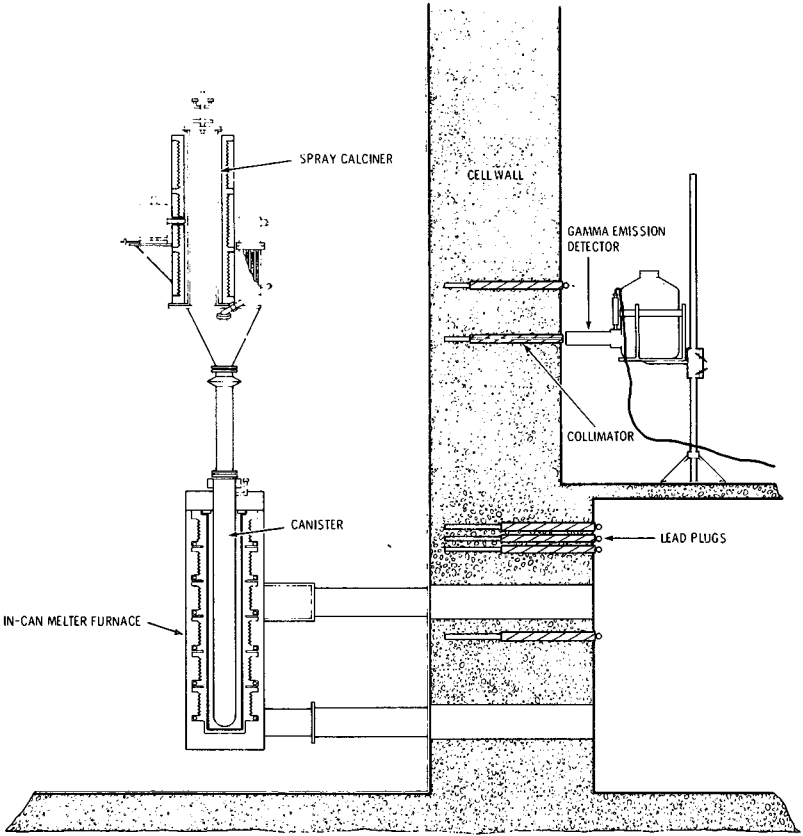


FIG. 3. In-Can Melter Level Detection System

In preparation for demonstrating the gamma emission monitoring technique, six horizontal holes were drilled partially through the cell wall. The holes were fitted with interchangeable lead collimators having bore diameters ranging between 0.25 to 3.2 mm. The collimators permitted the detector to "view" approximately 1 cm² of the canister while in the melter at four vertical locations. Collimators were also placed to allow viewing of the calciner filter chamber and the calciner cone directly beneath the spray nozzle. Gamma emissions from the waste passed through equipment, miscellaneous in-cell items, the stainless steel cell liner and approximately 5 cm of concrete before reaching the collimator and detector. The detector, a germanium diode, was approximately 5 m from the radiation source.

During operations, the melt level was readily determined by monitoring ¹³⁷Cs emissions. Representative data from the first test is shown in Fig. 4. The sharp rise in the count rate at time 0 is due to calcined high-level waste falling past the collimator line of site in the canister. The second sharp rise in the count rate at about 20 h represents the melt level reaching the height of the first collimator. This radiation increase by a factor of 10 in less than an hour provides accurate level indication. Similarly, other collimators higher up the canister allowed level detection at other heights.

A slight "overshoot" is apparent before the constant activity level of the final glass product is attained. This is because the bulk density of the calcine/frit mixture is lower than that of the glass, but as the calcine/frit layer melts the bulk density increases. The higher density provides internal shielding, slightly reducing radiation emissions.

Radionuclide Deposition Determination by Gamma Emission

Collimators viewing the calciner also provided valuable deposition data. Following the first run, very high gamma emission from the calciner cone directly below the spray nozzle indicated a buildup of dried waste in this area as a result of the previously discussed nozzle failure. Continuous monitoring at this point would have provided early warning of the buildup.

The activity level in the off-gas filter housing was monitored before, during and after the run. Since the spray calciner filters were operated at around 300°C during the test, selective deposition of Ru on the sintered stainless steel filters was anticipated. Such a deposition was observed. The concentrations of ¹³⁴Cs, ¹³⁷Cs, and ¹⁵⁴Eu jumped to 30% above cell background shortly after the start of the run and then remained constant

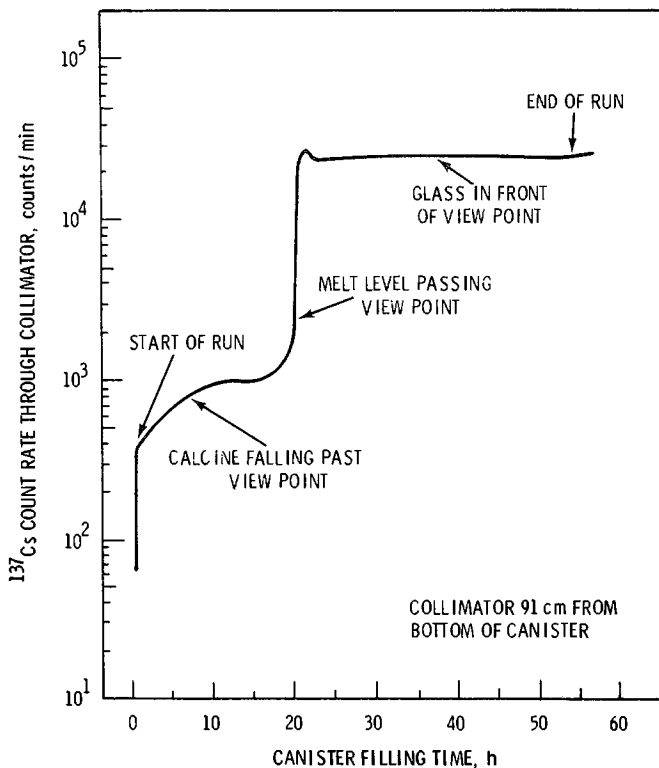


FIG. 4. Melt Level Indication by Gamma Emission

thereafter. The ^{106}Ru concentration, however, continued to increase throughout the course of the run until it reached a level 260% above cell background. Operation at higher temperatures as proposed for future spray calciners should minimize, if not eliminate, Ru deposition.

EFFLUENT DECONTAMINATION

Although determining the distribution of materials in the effluent system was not a goal of this project, a large number of samples from the effluent system were taken for analysis at a later date. A projected radiochemical balance based on the empirical values found in earlier operations of this effluent equipment is presented by Bjorklund and Hanson.⁵

Goles⁶ analyzed the NWVP process off-gas leaving the in-cell effluent treatment system prior to final filtration and discharge to the building stack. The effluent treatment system and the sample point used by Goles are shown in Fig. 5. The activities of particulates contained in the process off-gas are shown in Table VI. The activities of semi-volatile species in the gas stream are given in Table VII.

Data from monitoring the building stack is also available. Gross activities released during NWVP operations via the building stack are presented in Table VIII. Greater than 1.03×10^6 Ci of beta-gamma and 4.14×10^3 Ci of alpha materials were concentrated and solidified. Discounting the activity releases from all other sources in the building, the effluent system was shown to provide the decontamination factors^a given in Table IX. During vitrification and concentration, stack releases were well within allowable limits. Future analyses of the samples taken during operation will allow evaluation of the effluent decontamination efficiency of individual treatment steps. The total system, however, has been shown to be capable of adequately decontaminating effluents from vitrification of commercial HLW.

GLASS-FILLED CANISTER CHARACTERIZATION

To date, limited nondestructive examination of the NWVP glass has been performed. Both canisters are available in the PNL cells for further study and final disposition.

Radiation

Gross gamma emission, measured at a height 0.6 m from the bottom of Canister 1, was observed to be 4,080 rad/h 1 m from the canister and 1,500 rad/h at a distance of 2 m. Gamma emission

a. Decontamination factor is defined as the ratio of the activity of the material entering the process to that of the material exiting the process.

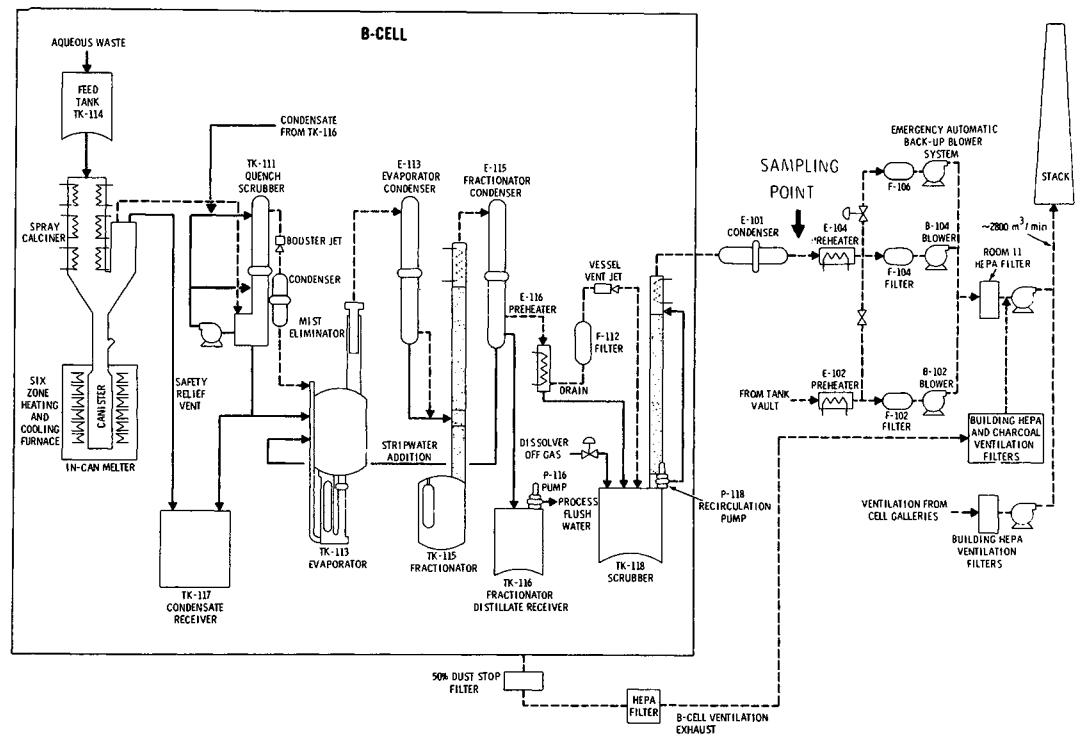


FIG. 5. Effluent Treatment System

TABLE VI. Radionuclide Content of Process Off-Gas Particulate Matter

<u>Isotope</u>	<u>Concentration,</u> <u>nCi/m³</u>
106Ru	44.0
125Sb	0.52
125mTe	1.7
134Cs	7.8
137Cs	52.0
144Ce	0.82
154Eu	0.28
155Eu	0.18
241Am	0.19

TABLE VII. Semi-Volatile Radionuclides in Process Off-Gas

<u>Isotope</u>	<u>Concentration</u>
79Se	20 fCi/m ³
99Tc	1.6 pCi/m ³
106Ru	200 nCi/m ³
125Sb	50 nCi/m ³
126Sb	3 nCi/m ³
125mTe	0.1 nCi/m ³
134Cs	6 nCi/m ³
137Cs	100 nCi/m ³
Total	10 ⁵ $\frac{\text{y/s}}{\text{m}^3}$

TABLE VIII. Gross Activity Released to Stack

Operation	Activity Released, Ci			Activity Concentration, Ci/cm ³		
	Alpha	Beta	Gamma	Alpha	Beta	Gamma
Run 1						
HLW Concentration ^a	1.25×10^{-7}	3.48×10^{-7}	b	1.56×10^{-1}	4.27×10^{-15}	b
SC/ICM ^a Operations	4.1×10^{-8}	4.6×10^{-8}	b	1.5×10^{-15}	1.6×10^{-15}	b
Run 2						
HLW Concentration ^a	1.7×10^{-8}	7.8×10^{-8}	b	0.4×10^{-16}	2.7×10^{-15}	b
SC/ICM Operations	1.8×10^{-8}	7.3×10^{-8}	1.64×10^{-3}	2.5×10^{-6}	7.7×10^{-16}	3.3×10^{-10}

a During spray calciner/in-can melter (SC/ICM) operations, the spent fuel dissolution system was in operation, contributing an undetermined amount to the activities shown.
 b Data not available.

TABLE IX. Effluent Decontamination Efficiency

Operation	Decontamination Factor	
	Alpha	Beta-Gamma
Run 1		
HLW concentration	7×10^7	a
SC/ICM ^b operations	2×10^{10}	a
Run 2		
HLW concentration	1×10^{11}	a
SC/ICM operations	7×10^{10}	2×10^8

(a) Data not available.

(b) Spray calciner/in-can melter.

from approximately 1 cm² along the canister axis was measured by Bryan³ during canister removal from the melting furnace. Relative concentrations of ¹³⁷Cs, ¹⁵⁴Eu and ¹⁰⁶Ru were measured as a function of axial position starting at the glass surface and working down to the bottom in 15 cm increments. The results are plotted in Fig. 6. The concentration of measured radionuclides appears to be relatively constant except for an unexplained enrichment of ¹⁰⁶Ru and ¹⁵⁴Eu and a depletion of ¹³⁷Cs approximately 15 cm below the glass surface.

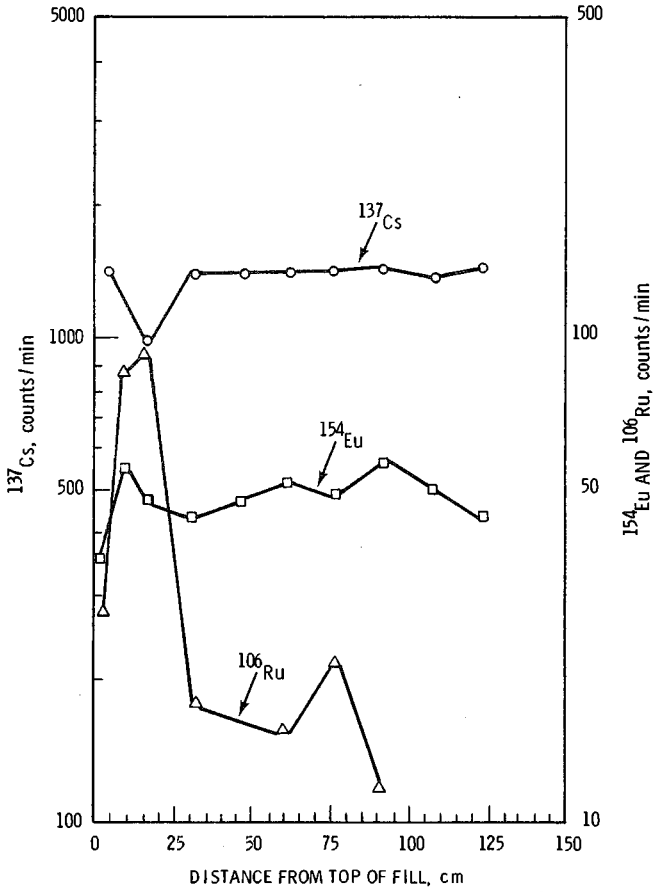


FIG. 6. Gamma Emission Scan of Canister 1

Thermal Properties

Surface and internal temperatures were measured by thermocouples on the canister exterior and in thermowells. Steady-state temperatures of Canister 1 hanging in air are shown in Fig. 7. Maximum centerline temperatures of 130°C were measured near the center of the filled portion of the canister. The maximum surface temperature was 103°C.

Heat output from Canister 1, as measured in a water bath calorimeter, was 350 W. From the heat generation rate and centerline-to-surface temperature differences, the apparent thermal conductivity of the glass is 1 W/m°C. This compares well with laboratory data.⁷ Apparently, internal cracking of the glass, which most likely occurs during cooldown, does not substantially decrease the thermal conductivity of the glass inside the canister.⁸

LABORATORY-SCALE IMMOBILIZATION TESTING

A portion of the commercial HLW produced for the engineering-scale vitrification demonstration was not consumed. This valuable waste will be used in a laboratory-scale immobilization system to obtain much needed effluent processing and waste form data. A laboratory-scale system (shown in Fig. 8) presently consisting of a spray calciner, an in-can melter and associated feed and off-gas systems has recently been installed in PNL hot cells. The equipment was designed in modules to readily accommodate alternative waste form processes. Because the laboratory-scale system operates at a nominal 1-L/h feedrate, many relatively long-term tests can be conducted with the available waste. Processing objectives are:

- evaluate effluents as a function of calciner and melter operating conditions and radioactive waste compositions
- test and improve effluent decontamination systems and evaluate a system similar to the proposed Savannah River Laboratory effluent control system
- produce solidified waste forms for evaluation
- provide a waste solidification system that can be routinely operated at minimum cost
- establish a system for radioactive testing of alternative waste forms.

Plans presently call for vitrification operations to begin in early 1980 with simulated, radioactive Savannah River Plant

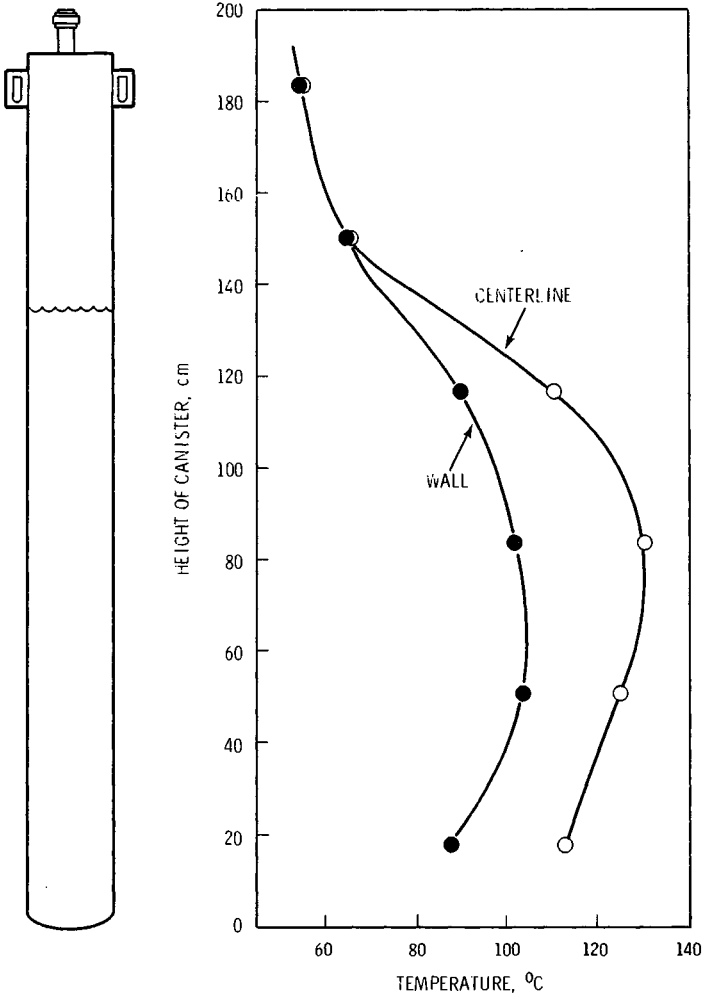


FIG. 7. Temperatures While Hanging in Air--Canister 1

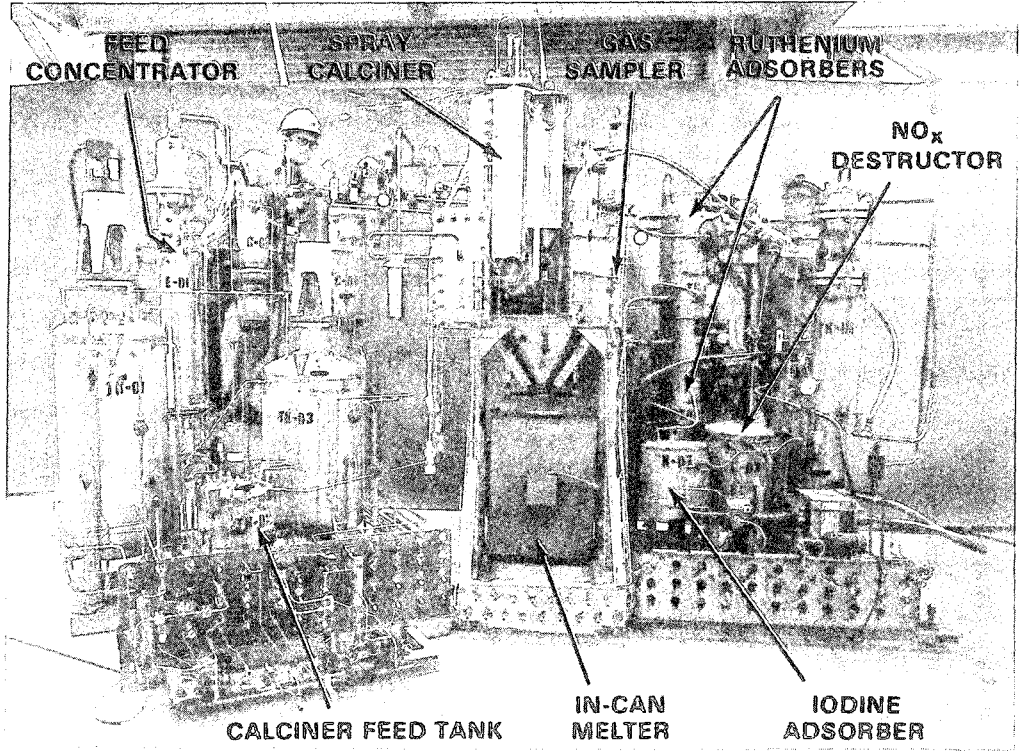


FIG. 8. Laboratory-Scale Immobilization Equipment

wastes produced using the existing HLW. Plans also include the vitrification and production of alternative waste forms with other HLW, such as West Valley, New York, waste and future commercial wastes.

CONCLUSIONS AND RECOMMENDATIONS

The NWVP demonstration was successful not only because of completing the objective of producing two canisters of vitrified, commercial HLW, but also because a large amount of technical information was and can be obtained from the demonstration. Because of NWVP, closing of the nuclear fuel cycle has been demonstrated on an engineering scale, showing that HLW can be immobilized safely and efficiently and advancing the national goal of energy independence. Several specific conclusions can be drawn from the NWVP demonstration. They are:

- High-level waste from reprocessing of commercial nuclear reactor fuel has been and can be converted to a solid, highly inert borosilicate glass on an engineering scale. Building on this available technology, the optimization and design of an operating facility can proceed.
- Gaseous effluents from the concentration, calcination and vitrification of HLW can be adequately decontaminated to allow their release to the atmosphere within current discharge limits.
- The spray calciner/in-can melter combination is a workable system for vitrifying HLW.
- High-level waste intrinsic gamma radiation can be used to determine waste levels in vessels and to monitor bulk and/or selective radiochemical depositions on processing equipment and piping.
- The two NWVP canisters of vitrified HLW exhibit predicted properties.

Due to the nature of NWVP, all obtainable data could not be gathered. Some recommendations for further study promising relatively high payoff per resources invested are:

- Effluent and liquid HLW samples obtained during NWVP operations should be analyzed and the results reported. These analyses can be used to determine radionuclide behavior during processing and the effectiveness of individual process equipment in controlling emissions.

- The two glass-filled canisters should be characterized. Non-destructive tests, including tests of thermal properties, radiation emissions, canister condition, etc, should be performed for both canisters. One of the canisters should be core-drilled in several places and samples removed for characterization of the only engineering-scale, commercial HLW glass in the nation.
- Small-scale testing with the HLW remaining from NWVP should be conducted and plans to do so are under way. In-cell laboratory-scale equipment will allow evaluation of effluent properties, waste form characteristics and processing parameters at a relatively low cost and yet representative scale.

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