QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
FIXATION OF RADIOACTIVE RESIDUES
NOVEMBER, DECEMBER, 1968 JANUARY, 1969

FEBRUARY 1969

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Edited by

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PREVIOUS QUARTERLY REPORTS

<u>Quarter</u>	<u>Year</u>	<u>Report Number</u>	<u>Reference*</u>
	<u>1959</u>		
First		HW-60584	59-1
Second		HW-13635	59-2
Third		HW-63048	59-3
Fourth		HW-63949	59-4
	<u>1960</u>		
First		HW-65209	60-1
Second		HW-66571	60-2
Third		HW-67334	60-3
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	<u>1961</u>		
First		HW-69176	61-1
Second		HW-70517	61-2
Third		HW-71374	61-3
Fourth		HW-72324	61-4
	<u>1962</u>		
First		HW-73412	62-1
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	<u>1964</u>		
First		HW-81862	64-1
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	<u>1965</u>		
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	<u>1966</u>		
First		BNWL-276	66-1
Second		BNWL-306	66-2
Third		BNWL-365	66-3
Fourth		BNWL-410	66-4

* Reference notation used in this report.

PREVIOUS QUARTERLY REPORTS

<u>Quarter</u>	<u>Year</u>	<u>Report Number</u>	<u>Reference*</u>
	<u>1967</u>		
First		BNWL-434	67-1
Second		BNWL-507	67-2
Third**		BNWL-677	67-3
Fourth		BNWL-771	67-4
	<u>1968</u>		
First		BNWL-834	68-1
Second		BNWL-889	68-2
Third		BNWL-923	68-3

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* Reference notation used in this report.

** Reports for the period July through October, 1967. Reporting periods are now February-April, May-July, August-October, and November-January.

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INTRODUCTION

This progress report is the fortieth in a series presenting research and development activities in the field of radioactive wastes.

Experimental work charged to programs other than those of the Division of Reactor Development and Technology is sometimes included for general interest and completeness; such work is identified in the headings.

SUMMARY

Spray solidification Run SS-6 was completed by reducing 986 liters of sulfate-free PW-1 waste to 65 liters of ceramic solid in 55.6 hr. The 3,030,000 Ci of radioactivity interfused in the solidified waste produced an internal heat generation rate of 10,800 W.

During spray solidification Run SS-7, 839 liters of PW-1 waste containing 3,620,000 Ci of radioactivity were reduced to 64 liters of ceramic solid in 47 hr. The solidified waste produced an internal heat generation rate of 12,700 W which represents the maximum internal heat generation rate in any WSEP receiver pot to date. The centerline and wall temperatures of the receiver pot while hanging in air were 928 and 427 °C, respectively, representing a thermal maximum in air.

A PW-4m phosphate feed suitable for spray solidification was developed and tested in the cold semiworks spray solidifier. It was necessary to dilute the feed to 585 liters/tonne to achieve adequate feed flow control. In two additional runs, ruthenium volatilization was shown to increase with feed acidity and essentially no ruthenium volatilized at a feed acidity near zero.

A radioactive WSEP waste pot was installed in an environmental test pod in the Solids Storage Engineering Test Facility (SSETF) to begin the interim storage studies. The first pot is from phosphate glass Run PG-4 and currently contains 2.2 kW of fission product decay heat.

SPRAY SOLIDIFICATION

LABORATORY STUDIES

Corrosion Studies - R. F. Maness

The freeze valve on the spray calciner melter has been modified to operate by passing air through Hastelloy X tubing wound around the outlet tube. Hastelloy X corrosion under these conditions was simulated by producing PW-2 spray calciner melt at 925 °C in an alumina crucible equipped with an overflow weir and allowing the melt to overflow into another alumina crucible also held at 925 °C and containing a Hastelloy X specimen in the vapor phase. Corrosion of the Hastelloy X specimen was essentially nil after 110 exposure hours (28 hr with semicontinuous feed addition).

Specimens of 304L and 310 stainless steel were exposed to PW-4m spray calciner melt at 925 °C for 115 hr. Corrosion rates were 46 and 13 mils/year, respectively. Similar specimens, as well as a 50 Cr-50 Ni alloy specimen, were exposed to PW-4m spray calciner melt at 1000 °C for 38 hr. The 304L (1/4 in. thick) and 310 (1/8 in. thick) specimens were consumed. The 50 Cr-50 Ni alloy exposed to PW-1 and to PW-2 spray calciner melts at 800 °C for 145 hr corroded at rates of 7 and 60 mils/year, respectively. A stainless steel cylinder flame sprayed with 50 Cr-50 Ni alloy (about 20 mil coating) was exposed to PW-2 spray calciner melt at 800 °C for 48 hr. The coating was found to be spalled when removed from the melt and cooled to room temperature.

ENGINEERING STUDIESDevelopment Spray Solidification Runs - J. D. Moore

A PW-4m phosphate feed suitable for spray solidification was developed and tested in the cold semiworks spray solidifier. It was necessary to dilute the feed to 585 liters/tonne to achieve adequate feed flow control. In two additional spray solidifier runs, ruthenium volatilization in the spray solidification process from PW-1 feed was shown to increase with feed acidity. At a feed acidity of zero, essentially no ruthenium was volatilized. Feed compositions for the three runs are listed in Table 1 and solidifier operating conditions and results are summarized in Table 2.

A. Run DSC-22

PW-4m was modified by the addition of 1.5M sodium, 1.2M iron, and 2.46M phosphoric acid (378 liters/tonne basis) to produce a feed which could be fused to a homogeneous solid. During a 9 hr run, this feed had to be diluted with water so that the pump could provide sufficient flow to the spray solidifier. An initial concentration of 470 liters/tonne was diluted in two steps to a final concentration of 585 liters/tonne before an adequate flow rate (14 liters/hr) could be maintained. The feed did not contain noticeable precipitates, but it was very dense (1.44 g/cm^3 at 585 liters/tonne) and became more viscous when agitated and circulated.

The calciner filters remained relatively clean with a blowback pressure of 40 psig. The filter pressure drop increased uniformly from 2.5 to 3.8 in. of water over the last 5 hr of the run. At the end of the run,

TABLE 1. *Feed Compositions for Development Spray Solidifier Runs (Basis: 378 liters/tonne concentration)*

Component	DSC-22	DSC-23	DSC-24		
Waste Type	PW-4m	PW-1	PW-1		
Fe ⁺³	0.124	0.955	0.946		
Cr ⁺³	0.012	0.012	0.012		
Ni ⁺²	0.008	0.005	0.005		
Al ⁺³	0.001	0.001	0.001		
ZrO ⁺²	0.143	0.065	0.065		
Na ⁺	0.342	0.178	0.138		
SiO ₃ ⁻²	0.010	0.010	0.010		
PO ₄ ⁻³	0.003	0.003	0.003		
MoO ₄ ⁻²	0.13	0.065	0.065		
Sr ⁺²	0.036	0.0155	0.0155		
Ba ⁺²	0.041	0.0195	0.0195		
Rb + Cs (used K ⁺)	0.092	0.042	0.042		
(Y + RE) ⁺³	0.274	0.12	0.12		
Ru ⁺⁴	0.0082	0.0064	0.016		
Rh ⁺² (used Co ⁺²)	0.013	0.0074	0.0074		
Ag ⁺	0.0016	0.0008	0.0008		
Cd ⁺²	0.0025	0.0008	0.0008		
TeO ₄ ⁻² (used SO ₄ ⁻²)	0.015	0.0064	0.0064		
Pd ⁺² (used Ni ⁺²)	0.043	0.017	0.017		
TcO ₄ ⁻²	0.031 (used MoO ₄ ⁻²)	0.014 (used MoO ₄ ⁻²)	0.014 (used Mn ⁺²)		
NO ₃ ⁻	6.6	3.6	3.63		
H ⁺³	4.8	0.022	0.022		
Additives					
	Batch #1	Batch #2	Batch #3		
Na ⁺	1.258	1.0	1.0	1.8 (a)	
Fe ⁺³	1.2	--	--	--	
PO ₄ ⁻³	2.46	0.69	1.015	1.34	--
H ⁺	7.38	2.07	4.315	6.57	--
NO ₃ ⁻	3.6	--	1.27	2.55	--
OH ⁻	1.258	--	0.325	0.65	--
PO ₃ ⁻	--	1.0	0.675	0.35	1.8 (a)

a. Added as solid to the melter.

TABLE 2. *Summary of Development Spray Solidification Runs*

	<u>DSC-22</u>	<u>DSC-23</u>	<u>DSC-24</u>
Feed type	PW-4m	PW-1	PW-1
Feed concentration, liters/tonne	470-585	454	378
Additives	$\text{Fe}^{+3}, \text{Na}^{+}, \text{PO}_4^{-3}$	$\text{Na}^{+}, \text{PO}_3^{-}, \text{PO}_4^{-3}$	NaPO_3 , solid frit
M ⁺ /P	2.75	2.9	3.0
Feed consumed, liters	125.5	105, part 1 80.5, part 2 74, part 3	181
Average feed, rate liters/hr	14	15.6	15
Calcliner furnace temperature, °C	700	700	700
Calcliner vacuum, in. of water	2-10	10	8
Atomizing gas	air	air	air
Atomizing gas flow, scfm	6	6	7
Filter blowback pressure, psig	30-40	40	32-40
Filter pressure drop, end of run, in. of water	3.75	5.75	5.5
Melter furnace temperature, °C	1100-1150	1150-1175	1150
Product receiver furnace temperature, °C	800	850	600
Product collected, kg	35.5	63	53.7
Product density, g/cm ³	3.5	3.6	3.3
Particulate DF (feed to con- densate)	1000	540	460
Product concentration, liters/tonne	50	31	34
Solid NaPO_3 added to melter, kg	0	0	34

the blowback pressure was increased to 60 psig for three blowback cycles. Less than 1/8 in. of powder coated the filters after the run.

Calcine deposits about 1/4 in. thick were found about 2 ft below the feed nozzle on the calciner wall. The deposits were probably formed early in the run when the feed rate was erratic. The rest of the calciner wall was normal with a coating of up to 1/8 in. of powder.

The melter furnace temperature was increased from 1100 to 1150 °C early in the run in order to prevent excessive powder accumulation in the melter. The melter was successfully emptied twice through the drain valve, and both the weir and drain valve could be sealed with the cooling air coils around them. With the melter at 1150 °C, the melt was quite viscous and could not be overflowed from the weir fast enough to prevent a continuous increase in level in the melter. The weir would have drip-discharged at a faster rate if some of the insulation around the outside of the wier cooling coil had been removed.

Melt was collected without stalagmite formation in the 304L stainless steel pot heated at 800 °C. A 1 ft long section of the pot internal thermowell was completely dissolved by the melt after a melt-batch was dumped into the pot. The pot wall was not noticeably corroded. The maximum pot centerline temperature ranged between 950 and 1000 °C for about an hour during this time. The thermowell corrosion rate was greater than 113 mils/hr. Later corrosion tests with the melt showed that the rate for 304L stainless steel was reduced to only 46 mils/year at 925 °C.

Melt product was a homogeneous, dark-brown, crystalline solid with a density of 3.5 g/cm^3 and a concentration of 50 liters/tonne.

For the duration of the run, less than 4 percent of the ruthenium was volatilized from the feed. During the last 2 hr of the run, however, volatilization increased twenty-fold so that about 17 percent of the total ruthenium was volatilizing when the run was ended. The nonuniform volatilization may have been the result of a nonhomogeneous feed.

B. Runs DSC-23 and 24

Runs DSC-23 and 24 were made to observe the effect of PW-1 feed acidity on volatilization of ruthenium from the feed in the spray solidification process. In DSC-23, simulated PW-1 feed with additives of Na^+ , PO_4^{-3} , and PO_3 was prepared in three batches with acidities ranging from 2.1 to 5.9M. Each batch was run for approximately 6 hr with identical spray solidifier operating conditions (see Table 2) while condensate, scrubber, calcine, and melt product samples were collected. In DSC-24, feed was made up to contain only 0.022M free acid as compared to a normal acidity of 3.7M (8.9M including melt-forming additives). The only melt-forming additive used in this run was sodium metaphosphate which was added as solid chips to the melter. Analyses of the condensate and caustic scrubber samples showed that ruthenium volatilization increased nearly linearly from less than 0.5 to 33 percent with feed acidity (see Figure 1). To verify the ruthenium data from liquid samples, noncondensable off-gases from the condenser were also treated during DSC-24. One SCFM (10 percent of total flow) from the noncondensable off-gas flow was continuously passed through a ruthenium trap consisting of formaldehyde-treated silica gel in series with a bed of small polyethylene rings. Both

FIGURE 1. Ruthenium Volatilization from PW-1 Feed in the Spray Solidifier

of these materials are effective absorbers of gaseous ruthenium tetroxide. At the end of the run, neither material contained a trace of black coloring which would indicate the presence of ruthenium dioxide deposits.

The spray solidifier operated smoothly throughout both runs. Inspection of the internal walls and cone of the calciner showed that no calcine deposits over 1/8 in. thick had formed. The off-gas filters stayed relatively clean with filter blowback pressures from 32 to 40 psig. The filter pressure drop at the end of the runs was relatively low at less than 6 in. of water. Off-gas particulate deentrainment factors of about 500 were obtained for both runs. In DSC-24, deentrainment improved by a factor of two during the first half of the run. A thin coating of powder which formed on the filters during this time increased their efficiency.

Melt was drip-discharged from the melter weir early in the runs, but the melt in the weir tip eventually froze and batch dumping through the drain valve was used thereafter.

The product was collected without stalagmite formation in heated 304L stainless steel pots. The solidified melt was a homogeneous, dark-gray, crystalline material with densities of 3.6 and 3.3 g/cm³ for DSC-23 and 24, respectively.

RADIOACTIVE DEMONSTRATION - W. R. Bond, J. H. Hartley, and A. S. Neuls

During the quarter, the eighteenth and nineteenth pilot plant demonstrations of high level radioactive waste solidification were successfully completed with spray solidification Runs SS-6 and SS-7. A total of 6,650,000 Ci of radioactivity were interfused in 129 liters of ceramic solid to produce a total internal heat generation rate of 23,500 W. Sulfate-free PW-1 waste with additives of LiOH, NaOH, and H₃PO₄ was processed. Tables 3 and 4 present a summary of data and feed compositions for each run.

The internal-mix atomizing nozzle used to spray the feed into the solidifier operated satisfactorily during the runs except on one occasion it had to be flushed to remove a plug and on other occasions the nozzle cleanout needle had to be actuated to remove partial plugs. The floating-type pneumatic vibrator on top of the calciner barrel performed satisfactorily at removing calcine from the calciner walls. During the runs the filter pressure drop ranged between 4 to 6 in. of water indicating that the blowback system performed satisfactorily. A new melter drain valve and weir cooling assembly* was used throughout the run. The melt in the discharge valves was solidified by an air stream. To discharge the melt, the cooling air was turned off and the heat from the melter furnace thawed the material in either the weir overflow or the drain

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* BNWL-923, 68-3

TABLE 3. Summary Data for WSEP Spray Solidifier Runs

Operational Data	SS-5	SS-6	SS-7
Solidifier Run/WSEP Run	5/17	6/18	7/19
Solidifier	Spray	Spray	Spray
Date	10/68	11/68	12/68
Operating Mode	A	A	A
Feed Type (a)	PW-1	PW-1	PW-1
Feed, liters/tonne	446	521	452
Pot Diameter, in.	8	8	8
Pot Material	304-L	304-L	304-L
Total Run Feed Time, hr	53.2	55.6	47
Average Feed Rate, liters/hr	16.7	17.7	17.9
Total Feed, liters	887	986	839
Total Feed, tonne equivalent	2.0	1.9	1.85
Feed: Solid Volume Ratio	13.3	15.6	13.1
Feed: Solid Volume Ratio at 378 liters/tonne Basis	11.4	11.3	11.0
Filter ΔP , in. of Water	6.0	6.0	6.0
<u>Problems</u>			
SS-5 Power lead to melter furnace lost insulation and began arcing; insulation replaced with 3 1/2 hr delay in run.			
SS-6 (1) Partial plugging of the calciner inlet to the melter required two shutdowns to remove calcine from this area. (2) Calciner vibrator failed and was replaced.			
SS-7 Feed nozzle plugged halfway through the run due to inadequate feed flow measurement.			
<u>Filled Pot Data</u>			
Total Curies in Feed to Pot (b)	2,500,000	3,030,000	3,620,000
Total Curies of Ru in Feed to Pot (b)	16,800	24,100	22,000
Solid in Pot, liters	66	65	64
Solid in Pot, kg (c)	214	207	206
Solid in Pot, liters/tonne	33	35	34
Heat in Pot, W (d)	8200	10,800	12,700
Equivalent Aging Time, years	1.1	0.73	0.55
$\bar{\epsilon}$ Temp in Air-Cooled Furnace, °C	633	767	928
$\bar{\epsilon}$ to Wall Δ Temp., °C	278	308	450
Effective Thermal Conductivity, W/(m ²) (°C/m) [Btu/(hr) (ft ²) (°F/ft)]	1.1 [0.64]	1.39 [0.79]	1.1 [0.64]
Welded Lid	Yes	Yes	Yes
Radiation Reading, R/hr at 6 in.	Na	5.1 x 10 ⁴	NA

NA Not Available.

a. See Table 2 for feed composition.

b. By feed analysis.

c. Calculated from lab density and rodded volume

d. By pot calorimetry data.

TABLE 4. PW-1 Feed Compositions for Spray Solidification Runs SS-5, SS-6, and SS-7

Component	SS-5		SS-6		SS-7	
	Molarity, M(a)	Lab Analysis, M(a)	Molarity, M(a)	Lab Analysis, M(a)	Molarity, M(a)	Lab Analysis, M(a)
H ⁺	--	5.27	--	6.3	--	
Fe ⁺³	0.793 (b)	0.75 (b)	0.797 (b)		0.792 (b)	0.69 (b)
Cr ⁺²	0.012		0.012		0.012	
Ni ⁺²	0.005		0.005		0.005	
Al ⁺³	0.137 (b)	0.19 (b)	0.133 (b)	0.105 (b)	0.137 (b)	
Na ⁺	0.138	0.67 (c)	0.138	0.596 (c)	0.138	0.58 (c)
U ⁺⁶	0.01		0.01		0.01	
NO ₂ -NO ₃ ⁻	7.5	6.4 (d)	7.5	6.7 (d)	7.5	4.7 (d)
SO ₄ ⁻²	--		--		--	
PO ₄ ⁻³	0.003		0.003		0.003	
SiO ₂ ⁻²	0.01		0.01		0.01	
Cs ⁺ (added K ⁺)	0.0416		0.0416		0.0416	
Sr ⁺²	0.0155		0.0155		0.0155	
Ba ⁺²	0.0195		0.0195		0.0195	
(Y + RE) ⁺³	0.12		0.12		0.12	
ZrO ⁺²	0.065		0.065		0.065	
MoO ₄ ⁻² + Tc as MoO ₄ ⁻²	0.079		0.079		0.079	
Ru ⁺⁴	0.0032		0.0032	0.0039	0.0032	0.0039
Ru ⁺⁴ (added Fe ⁺³)	0.0288		0.0288		0.0288	
Rh ⁺² (added Co ⁺²)	0.0074		0.0074		0.0074	
Pd ⁺² (added Ni ⁺²)	0.0222		0.0222		0.0222	
Ag ⁺ + Cd ⁺² (added Cu ⁺²)	0.0016		0.0016		0.0016	
TeO ₄ ⁻² (added SO ₄ ⁻²)	0.0064		0.0064		0.0064	
Additives to Feed						
Li ⁺ (as hydroxide)	0.5		0.5		0.5	
Na ⁺ (as hyaroxiue)	0.5		0.5		0.5	
PO ₄ ⁻³ (as H ₃ PO ₄)	1.713	1.67	1.732	1.99	1.778	1.60
Final Feed						
Volume/tonne		466		521		452
Waste Equivalents						
EM _T ⁺	4.70		4.74		4.89	
EM _T ⁺ /P	2.75		2.75		2.75	
Sp Gr at 25 °C		1.36		1.32		1.31
Vol% Solids		~30		~20		~36
Total Radioactivity, Ci/liter ^(e)		2260		3070		4310
Radioruthenium, Ci/liter ^(e)		18.9		24.2		26.2
Total heat, W/liter ^(e)		9.27		11.3		15.3

a. Molarities are at 378 liters/tonne basis.

b. Excess aluminum in 1WW wastes replaces part of iron (including 0.0288M Fe substituted for ruthenium).

c. Includes sodium from additive.

d. Includes nitrate from additives.

e. By feed analysis.

valve. The melter was batch dumped via the drain valve in 5 to 6 liter batches every 4 to 6 hr. The melter would begin to drain about 2 min after the air was turned off to the cooling coils around the drain valve. The batch dumping of the melts allowed uniform filling of the receiver pots which were unheated with exception of the first half of Run SS-5.

Spray Solidification Run SS-5

During Run SS-5, waste equivalent to 2.0 tonnes of power reactor fuel (20,000 MWd/tonne at 15 MW/tonne) with an equivalent aging time of 1.1 years was processed to produce 214 kg of ceramic product. The 66 liters of radioactive ceramic produced a heat-rate density of 125 W/liter (8200 W total based on analytical data and pot calorimetry) in an 8 in. diameter 304L stainless steel melt receiver. The steady-state centerline temperature difference was 278 °C. These values indicate an effective thermal conductivity of 1.1 W/(m²)(°C/m) [0.64 Btu/(hr)(ft²)(°F/ft)] for the ceramic product.

The solidifier was fed 887 liters of adjusted sulfate-free PW-1 feed (additives of 0.5M LiOH, 0.5M NaOH, and 1.7M H₃PO₄ based on 378 liters/tonne) at 446 liters/tonne in 53.2 hr at an average feed rate of 16.7 liters/hr. The feed containing 2,500,000 Ci of radioactivity including 17,000 Ci of radioactive ruthenium was reduced in volume by a factor of 13.3, or 11.4 on a 378 liters/tonne basis.

The distribution of the feed radioactivity for Runs SS-5, SS-6, and SS-7 is given in Table 5. During Run SS-5, 70 percent of the ruthenium in the feed to the solidifier was volatilized

TABLE 5. Activity Distribution for Runs SS-5, SS-6, and SS-7 (Mode A Operation)*

	Fraction of Radioactivity in Condensate								
	¹⁰⁶ Ru			¹⁴⁴ Ce-Pr			Total Less Ru		
	SS-5	SS-6	SS-7	SS-5	SS-6	SS-7	SS-5	SS-6	SS-7
Through Process Condenser (E-111)	7 ⁻¹ ^(a)	4.6 ⁻¹	6.1 ⁻¹	6.3 ⁻⁴	6.4 ⁻⁴	2.6 ⁻²	6.3 ⁻⁴	6.4 ⁻⁴	2.6 ⁻²
Through Evaporator Condenser (E-113)	6.4 ⁻³	6.4 ⁻³	1.3 ⁻²	4.8 ⁻⁷	NA	2.8 ⁻⁵	4.8 ⁻⁷	NA	2.8 ⁻⁵
Through Fractionator Condenser (E-115)	6.0 ⁻⁵	2.0 ⁻⁵	2.8 ⁻⁶	3.0 ⁻¹⁰	NA	4.7 ⁻¹⁰	3.0 ⁻¹⁰	NA	4.7 ⁻¹⁰
Off-Gas to Scrubber	8.5 ⁻⁵	3.6 ⁻⁵	5.1 ⁻⁵	1.7 ⁻⁸	NA	NA	1.7 ⁻⁸	NA	NA
Off-Gas to Stack	<2.8 ⁻¹⁰	<2.8 ⁻¹⁰	<2.8 ⁻¹⁰	8.9 ⁻¹³	5.3 ⁻¹³	3.6 ⁻¹³	8.9 ⁻¹³	5.3 ⁻¹³	3.6 ⁻¹⁴
Ratio (Fract. Cond.) Activity to 10CFR20	1.8 ⁺⁵	2.1 ⁺⁴	1.0 ⁺⁴	1.1 ⁺³	5.1 ⁺³	2.2 ⁺²			
Ratio (stack) Activity to 10CFR20	<4 ⁻⁴	<4 ⁻⁴	<4 ⁻⁴	3.1 ⁻²	1.6 ⁻³	1.6 ⁻²			

NA Not Available

* BNWL-507, 67-2

a. 7⁻¹ = 7 x 10⁻¹

from the solidifier, while only 0.06 percent of all other combined radioactive species were entrained. Figure 2 shows the ruthenium accumulation in the WSEP evaporator during Runs SS-5, 6, and 7.

FIGURE 2. Ruthenium Accumulation in the WSEP Auxiliary Evaporator

The calciner condensate averaged 3.34M nitrogen throughout the run. This amount accounts for only 60 percent of the total nitrogen volatilized from the calciner. The remaining 40 percent remained as oxides of nitrogen, part of which were carried on through the WSEP evaporator and fractionator. This resulted in the scrubber solution being neutralized from 2M to 0.04M NaOH at the end of the run.

The cumulative decontamination factor (DF_c) for ruthenium across the WSEP auxiliary evaporator, was 1.1×10^2 . Instantaneous ruthenium DF_i 's ranged from 2.0×10^2 when feed was on

to 1.4×10^4 when feed was off. A nearly constant buildup rate of ruthenium in the acid fractionator and a higher than expected ruthenium concentration in the fractionator indicated a direct gas-phase flow of ruthenium from the process condenser to the evaporator condenser. Cumulative DF_c for $^{144}\text{Ce-Pr}$ representing nonvolatiles was 1.5×10^3 across the evaporator. Instantaneous DF_i 's averaged greater than 10^3 .

The DF_c across the fractionator for ruthenium was 1.2×10^2 and for $^{144}\text{Ce-Pr}$ was 3.2×10^3 . Instantaneous DF_i 's for ruthenium averaged greater than 10^2 and agreed with the cumulative value.

The final condensate stream (fractionator condensate) contained a fraction of solidifier feed activity equal to 5.9×10^{-5} for ruthenium and 3×10^{-10} for nonvolatiles. These values were above 10CFR20 release limits by factors of 1.8×10^5 and 1.1×10^3 , respectively. The large value for ruthenium also indicates the gas-phase loss of ruthenium described above.

Fractional losses of activity by off-gases to the stack were equal to less than 2.8×10^{-10} for ruthenium and 8.9×10^{-13} for gross beta. These values are below the 10CFR20 release limits by factors of $<4 \times 10^{-4}$ and 3.1×10^{-2} , respectively.

Spray Solidification Run SS-6

During Run SS-6, waste equivalent to 1.9 tonnes of power reactor fuel (20,000 MWd/tonne at 15 MW/tonne) with an equivalent aging time of 0.73 years was processed to produce 207 kg of ceramic product. The 65 liters of radioactive ceramic produced a heat-rate density of 168 W/liter (10,800 W total based on analytical data and 11,100 W total based on pot calorimetry) in an 8 in. diameter 304L stainless steel melt receiver. The

steady-state pot centerline temperature in the air-cooled furnace was 767 °C and the centerline-to-wall temperature difference was 308 °C. These values calculated out to give an effective thermal conductivity of $1.4 \text{ W/(m}^2)(^\circ\text{C/m)}$ [$0.79 \text{ Btu/(hr)(ft}^2)(^\circ\text{F/ft)}$] for the ceramic product.

The solidifier was fed 986 liters of adjusted sulfate-free PW-1 feed at 521 liters/tonne in 55.6 hr at an average feed rate of 17.7 liters/hr. The feed containing 3,030,000 Ci of radioactivity including 24,100 Ci of radioactive ruthenium was reduced in volume by a factor of 15.6, or 11.3 on a 378 liters/tonne basis.

The floating-type pneumatic vibrator failed after 28.6 hr of run time and was replaced. The vibrator was successful in removing the powder from the calciner barrel. In the cone, however, a powder buildup led to a blockage of the melter inlet on two occasions, after 1 and 16 hr of run time. The cone was mechanically cleared by way of the solids-feeder port.

After the run, the melter was disconnected from the spray calciner and inspected to determine the cause of the plugging. Considerable buildup of melt was found in the melter inlet which caused a reduction in the melter inlet cross-sectional area of approximately 50 percent. The buildup was attributed to sparging of the melter through the drain valve just prior to the start of the run. A melter viewing port also became plugged with melt at this time and had to be rodded out. The melt buildup was slumped from the melter inlet by insulating the top of the melter and heating it to 1200 °C. This situation can be avoided in the future by assuring that the drain valve is sealed prior to running the melter at increased vacuum and temperature. A calcine deposit was also found in the

conical bottom section of the calciner directly below the barrel. A total of 10 to 12 kg of caked calcine was removed from this area. The deposition had probably accumulated over the last five runs (the solidifier was flushed prior to SS-2) as a result of inadequate feed atomization caused by intermittent off-standard conditions in feed flow. It should be noted that over the last five runs a total of 240 hr of feed-on time has been accumulated on the equipment and a total of 850 kg of ceramic product has been produced. Inspection of the calciner barrel and filters revealed them to be relatively clean. The spray nozzle was removed and replaced with a new one not only in view of the deposition in the calciner but also based on its increased air consumption (approximately 20%) apparently caused by internal erosion of the nozzle.

During the entire run, 47 percent of the ruthenium in the feed to the solidifier was volatilized from the solidifier, while only 0.058 percent of the other combined radioactive species were entrained.

Nitrogen analysis of the calciner condensate accounted for only 75 percent of the total nitrogen volatilized from the solidifier. The remaining 25 percent remained as oxides of nitrogen which were carried through the WSEP evaporator and fractionator in the gas phase as in Run SS-5. This situation resulted in the scrubber solution being continuously neutralized from 2M NaOH at the start of the run to 0.07M NaOH at the end of the run.

The cumulative decontamination factor, DF_c , for ruthenium across the WSEP auxiliary evaporator was 7.3×10^1 . Instantaneous decontamination factors, DF_i 's, for ruthenium ranged from 1.5×10^2 near the start of the run to 2.7×10^6 after the solidifier feed was off. This, combined with a nearly

constant and excessive rate of ruthenium accumulation in the acid fractionator indicated direct gas phase flow of ruthenium from the process condenser to the evaporator condenser as occurred in Run SS-5. The DF_i 's across the evaporator for nonvolatiles represented by $^{144}\text{Ce-Pr}$ averaged greater than 10^4 .

The DF_c across the fractionator was 3.1×10^2 for ruthenium. The DF_i 's for ruthenium averaged greater than 8.0×10^2 .

The final condensate stream (fractionator condensate) contained a fraction of original feed activity equal to 2.0×10^{-5} for ruthenium and 8×10^{-9} for nonvolatiles. These values were above 10CFR20 release limits by factors of 2.1×10^4 and 5.1×10^3 .

Fractional losses of activity by off-gases to the stack were less than 2.0×10^{-10} for ruthenium and equal to 5.3×10^{-13} for gross beta. These values were below the 10CFR20 release limits by factors of $<4 \times 10^{-4}$ and 1.6×10^{-3} , respectively.

Spray Solidification Run SS-7

During Run SS-7, waste equivalent to 1.85 tonnes of power reactor fuel (20,000 MWd/tonne at 15 MW/tonne) with an equivalent aging time of 0.6 years was processed to produce 200 kg of ceramic product. The 64 liters of radioactive ceramic produced a heat rate density of 198 W/liter (12,700 W total based on analytical data) in an 8 in. diameter 304L stainless-steel melter receiver. The steady-state pot centerline temperature in the air-cooled furnace was 928 °C and the centerline-to-wall temperature difference was 450 °C. These values give an effective thermal conductivity of $1.1 \text{ W/(M}^2)(\text{°C/m})$ [$0.64 \text{ Btu/(hr)(ft}^2)(\text{F/ft})$] for the ceramic product. The 12.7 kW represent the maximum heat generation rate in

any WSEP receiver pot to date, and centerline and wall temperatures while hanging in air in excess of 900 and 427 °C, respectively, represent a thermal-maximum pot in air.

The solidifier was fed 839 liters of adjusted sulfate-free PW-1 feed at 452 liters/tonne in 47 hr at an average feed rate of 17.0 liters/hr. The feed containing 3,620,000 Ci of radioactivity including 22,000 Ci of radioactive ruthenium was reduced in volume by a factor of 13.1, or 11.0 on a 378 liters/tonne basis.

Difficulty was experienced during the first 23 hr of the run with feed control as the magnetic flowmeter was inoperable and manual feed control was required. The atomizing feed nozzle became plugged as a result and a 4 hr shutdown was required to unplug it by flushing it from the air side with acid. The flowmeter was made operable during the shutdown and the last 24 hr of the run were completed trouble-free using automatic feed control.

During the entire run, about 61 percent of the ruthenium in the feed to the solidifier was volatilized from the solidifier, while 2.69 percent of the other combined radioactive species were entrained from the solidifier. The high entrainment was forty times more than in Runs SS-5 and SS-6 and can be partly attributed to a feed line flush which occurred after 24 hr of feed-on time (activity in the feed line was flushed to the evaporator). Further evaluation will have to be made to determine the actual entrainment value.

The cumulative decontamination factor, DF_c , across the WSEP auxiliary evaporator for ruthenium was 52. Instantaneous DF_i 's ranged from 5.1×10^2 to 1.3×10^3 . Again, as in previous runs, a nearly constant buildup of ruthenium in the acid

fractionator indicated a direct gas phase flow of ruthenium from the process condenser to the evaporator condenser. Cumulative DF_c for $^{144}\text{Ce-Pr}$ representing nonvolatiles was 9.8×10^2 across the evaporator. Instantaneous DF_i 's ranged from 2.5×10^3 to 4.1×10^4 .

The DF_c across the fractionator for ruthenium was 4.8×10^3 and for $^{144}\text{Ce-Pr}$ was 5.8×10^3 . Instantaneous DF_i 's for ruthenium averaged greater than 10^3 and agreed with the cumulative value.

The final condensate stream contained a fraction of original feed activity equal to 2.9×10^{-6} for ruthenium and 4.7×10^{-10} for $^{144}\text{Ce-Pr}$. These values were above 10CFR20 release limits by factors of 1×10^4 and 2.2×10^2 , respectively.

PHOSPHATE GLASS SOLIDIFICATION

LABORATORY STUDIES

Corrosion Studies - R. F. Maness

A PW-1 phosphate glass melt was prepared from an acid waste containing 0.03M fluoride. A 304L specimen was exposed to this melt at 800 °C and to a similar melt prepared from fluoride-free waste solution. The corrosion rates obtained were essentially identical, 11 and 12 mils/year, respectively.

STUDIES ON AUXILIARIES TO SOLIDIFICATION SYSTEMS

Highlights in Spray Solidifier Runs SS-5, SS-6, and SS-7 - P. C. Owzarski

Excellent data obtained on ruthenium buildup in the acid fractionator indicate that the buildup rate was constant

during each run. This virtually proved that 1 percent of ruthenium volatilized in the solidifier was bypassing the evaporator bottoms via the 20 scfm off-gas stream. Each process condenser was 99 percent effective in scrubbing ruthenium (most likely as RuO_4). This was not adequate for ruthenium control since nearly one Ci of ruthenium was detected in spent caustic scrub solutions. Steam added to the process condenser did not increase the scrubbing efficiency, which indicated that the tube walls were already wetted by process condensate. A full cone spray nozzle is being installed in the process condenser to effect better RuO_4 scrubbing. The spray will provide additional surface for scrubbing and will break up the laminar gas flow pattern in the condenser tubes.

During Run SS-6, sugar was added to the evaporator at 0.79 moles/hr. However, since the gaseous ruthenium was already too high, the volatilization step during evaporation was masked. At the end of the run, with feed to the solidifier off, ruthenium DF's obtained were from 2×10^5 to 2×10^6 .

Mathematic Model of Ruthenium Scrubbing - P. C. Owzarski and R. C. Arnett

An analog computer solution to the diffusion equation with absorption at the tube walls was carried out for laminar gas flow in the process condenser. Results showed that a gas phase diffusion limiting model underestimated the experimental scrubbing efficiency only slightly. This confirmed the relatively poor scrubbing effectiveness of the process condenser. An empirical equation for the scrubbing efficiency was obtained for tubes longer than 5 ft. It was assumed that RuO_4 is very dilute, so the wall concentration is effectively zero. The equation is:

Fraction Ru remaining in gas at 25 °C = $0.00146 V_{\max} R / (ZD) - 0.1$
where

Z = tube length, ft

R = tube diameter, ft

D = diffusivity, ft²/sec

$V_{\max}$ = centerline velocity, ft/sec.

DEVELOPMENT STUDIES OF SOLIDIFICATION PROCESSES

HEAT TRANSFER STUDIES

Estimation of Molten Core Size in Waste Storage Vessels and Frozen Wall Waste Matters - J. D. Kaser

Equations were derived for calculating the size of molten cores in cylindrical and spherical vessels containing highly radioactive heat-generating waste. These equations may be used for cases where (1) all heat is generated by radioactive decay, and (2) internal heaters supply heat in addition to the radioactive decay heat.

The equations were derived from simple idealizations and are therefore intended for preliminary sizing and evaluation of molten core storage vessels and frozen wall melters.

The following simplifying assumptions were used in the derivations:

1. The radioactive decay heat generation is uniform.
2. The thermal conductivity is uniform and independent of temperature in the solid portion of the waste.
3. The outer surface temperature of the vessel is uniform.
4. All temperatures are at their steady-state values.

5. The cylindrical vessels have insulated ends.
6. Any internal heaters are centrally located.
7. The heat transfer coefficient between the molten and solid waste is uniform, which means there are not thin spots in the frozen wall.

Storage pot calculations without heaters have been reported in graphical form by W. V. DeMeir.* A figure included at that time on pot wall temperature estimates was in error and is presented here in its correct form as Figure 3.

FIGURE 3. Pot Wall Temperatures in Air

* BNWL-507, 67-2

Cylindrical Vessels. At the boundary between the solid waste and molten waste, $r = r_m$, the radius of the molten zone, and $T_w = T_m$, the melting point of the waste. The relationship between r_m and T_m for a heat generating material with additional axial heating is:

$$\frac{4(T_m - T_o)k_w}{Ar_i^2} = 1 - \left(\frac{r_m}{r_i}\right)^2 - \left(\frac{2B}{\pi L r_i^2 A}\right) \ln\left(\frac{r_m}{r_i}\right) + \frac{2k_w}{k_c} \left(1 + \frac{B}{\pi L r_i^2 A}\right) \ln\left(\frac{r_o}{r_i}\right) \quad (1)$$

Equation (1) cannot be solved explicitly for (r_m/r_i) when both A and B have nonzero values. For the special case of storage vessels with no additional axial heating ($B = 0$), Equation (1) can be solved explicitly for (r_m/r_i) :

$$\frac{r_m}{r_i} = \left[1 - \frac{4(T_m - T_o)k_w}{Ar_i^2} + \frac{2k_w}{k_c} \ln\left(\frac{r_o}{r_i}\right)\right]^{1/2} \quad \text{when } B = 0 \quad (2)$$

Spheres. The relationship between r_m and T_m for a spherical container of heat generating material with additional central heating is:

$$\left(\frac{r_m}{r_i}\right)^3 + \left[\frac{6k_w(T_m - T_o)}{r_i^2 A} - 1 + \frac{3B}{2\pi r_i^3 A} - \frac{k_w}{k_c} \left(\frac{r_o - r_i}{r_o}\right) \left(2 + \frac{3B}{2\pi r_i^3 A}\right)\right] \times \left(\frac{r_m}{r_i}\right) - \frac{3B}{2\pi r_i^3 A} = 0 \quad (3)$$

Equation (3) can be solved for (r_m/r_i) using any standard method for cubic equations, however, this equation can be simplified further for two interesting limiting cases.

For a nonheat generating material, Equation (3) simplifies to:

$$\frac{r_m}{r_i} = \left[\frac{4\pi k_w r_i (T_m - T_o)}{A} + 1 - \frac{k_w}{k_c} \left(\frac{r_o - r_i}{r_o} \right) \right]^{-1} \quad (4)$$

While for a heat-generating material without additional internal heating Equation (3) can be simplified to:

$$\frac{r_m}{r_i} = \left[1 + \frac{2k_w}{k_c} \left(\frac{r_o - r_i}{r_o} \right) - \frac{6k_w (T_m - T_o)}{r_i^2 A} \right]^{1/2} \quad (5)$$

Equation (5) for a sphere is analogous to Equation (2) for a cylinder.

Examples of Applications

Sizing a Frozen Wall Melter. In this example, we will estimate the size of a spherical melter with a 1 in. thick frozen wall (1 in. of solidified waste) for processing waste with a specific heat generation rate of 15,000 Btu/(hr)(ft³) [155 W/liter]. The melter wall material is 1/2 in. thick and the outer surface is to be kept at 600 °F (316 °C). The thermal conductivity of the wall material is 5 Btu/(hr)(ft²) (°F/ft) [8.6 W/(m²)(°C/m)], and the thermal conductivity of the frozen waste is 0.5 Btu/(hr)(ft²) (°F/ft) [0.86 W/(m²)(°C/m)]. The melting point of the waste is 1380 °F (749 °C).

To size the melter, we first determine the maximum diameter that the sphere can have and still maintain a frozen wall 1 in. thick. Using Equation (5), we solve for r_i while expressing r_m as $r_i + a$ and r_o as $r_i + b$ where a and b are the thickness of frozen wall and the thickness of melter wall, respectively. Equation (5) rearranges into the following quadratic form:

$$\left(2a + \frac{2k_w b}{k_c}\right) r_i^2 + \left[2ab - a^2 - \frac{6k_w (T_m - T_o)}{A}\right] r_i - \left[a^2 b + \frac{6k_w b (T_m - T_o)}{A_o}\right] = 0 \quad (6)$$

Inserting numerical values, we find that $r_i = 0.933$ ft is the maximum value for the radius of the frozen wall melter.

Because of probable variation in the radioactivity and thermal properties of the waste, the radius of the actual melter should be less than the calculated radius. It will be assumed that experience suggests reducing the radius 10 percent, which means about a 30 percent reduction in volume and radioactive decay heat load. The practical melter diameter is estimated to be 20 in. Now the problem is to size the internal heaters needed to maintain a frozen wall thickness of 1 in. Therefore, we solve Equation (3) for B, the heater power, and find that the internal heater power must be at least 4000 Btu/hr (1172 W) in order to keep the frozen wall 1 in. thick.

Size of Molten Core in Cylindrical Storage Containers.

Here we will determine the molten core size when the heat generation is just great enough to keep the surface temperature of the container at 842 °F (450 °C), which is considered a safe limit for air storage.

Description of a container of waste:

Fill depth:	6 ft
Internal diameter:	0.667 ft
Thermal conductivity of waste:	0.5 Btu/(hr)(ft ²)(°F/ft) (0.86 W/(m ²)(°C/m)

Thermal conductivity of container

wall: $9 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F}/\text{ft})$
 $(15.6 \text{ W}/(\text{m}^2)(^\circ\text{C}/\text{m}))$

Melting point of waste: 1380°F (749°C)

Wall thickness: 0.3 in.

From Figure 3 it is estimated that the container will lose heat to the surroundings at a rate of $60,000 \text{ Btu/hr}$ ($17,580 \text{ W}$) when the surface temperature of the container is 842°F (450°C).

This corresponds to a volumetric heat generation rate of $\frac{9(60,000)}{6\pi} = 28,648 \text{ Btu}/(\text{hr})(\text{ft}^3)$ [279 W/liter].

From Equation (2) we find that $(r_m/r_i) = 0.818$. Thus, the radius of the molten core is 3.27 in. or the frozen wall thickness is 0.73 in.

Nomenclature

Variables.

A = volumetric heat generation rate, $\text{Btu}/(\text{hr})(\text{ft}^3)$

a = thickness of frozen waste, ft

b = thickness of vessel wall, ft

B = power of axial or central heater, Btu/hr

c = constant of integration

k = thermal conductivity, $\text{Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F}/\text{ft})$

L = length of filled portion of storage vessel, ft

r = radius of cylinder or sphere, ft

T = temperature, $^\circ\text{F}$

π = $3.1415\dots$, mathematical constant

Subscripts

c = container wall material
i = inner surface of vessel
m = molten zone boundary
o = outer surface of vessel
w = solid radioactive waste

Filling of Pots with Thermal Maximum Heat Content in Water -

J. L. McElroy

It has been concluded that thermal maximum pots intended for water storage* can be filled and safely handled in a plant that is designed to insure water cooling of the pot during filling, pot transfers, and interim storage. The amount of heat generating fission products that can be safely handled in a pot is dependent on product thermal conductivity, product melting or remelt temperature, melt corrosiveness, pot geometry and pot environment during filling and interim storage. Indirect factors that must also be considered include product stability and fission product migration. More will be learned later about two factors in long term product storage tests. It is not considered that any major problems will arise from filling pots while they are sitting in water.

Equations have been derived elsewhere in this report for calculating frozen wall thicknesses in pots with molten cores. Graphs were also presented in an earlier report for this same purpose.** One case is presented here to illustrate the type

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* Thermal maximum in water is the quantity of radioactive decay heat per unit volume of solidified waste which will produce a centerline temperature of 900 °C when the pot is stored in water at 100 °C. Thermal maximum pots with molten cores are based on extrapolation of the thermal conductivity of the waste in the solid state to the hypothetical centerline temperature of 900 °C.

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of considerations given to filling a pot with a high quantity of fission product heat. For cylindrical containers filled to a depth of 6 ft, Figure 4 shows the heat generation rate required at various product thermal conductivities in order for the pot centerline to attain a 900 °C centerline temperature. Figure 5 gives the product frozen wall thickness as a function of remelt temperature. Figure 5 is for an 8 in. diameter pot containing waste with a heat rate content equal to the thermal maximum in water limit for a centerline temperature of 900 °C taken from Figure 4. As an example, suppose a thermal maximum in water pot (hypothetical 900 °C centerline) is desired for a product with a thermal conductivity of $0.7 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F}/\text{ft})$. From Figure 4 the heat generation rate required is 23 kW. If the product has a melting point of 700 °C, the frozen wall thickness during water storage would be 2.0 in. (Figure 5). If the pot lost cooling water but had the benefit of natural convection cooling, the frozen wall thickness would decrease to about 1/2 in. Obviously, the later situation would give some cause for concern and some knowledge about the corrosiveness of the melt and the integrity of the frozen wall would be essential.

Similar examples using centerline temperatures either less than or greater than 900 °C could be discussed, but in the final analysis each particular case will have to be evaluated according to its own characteristics by considering the items listed above.

FIGURE 4. Heat Required in a Cylindrical Pot to Attain a 900 °C Centerline Temperature

FIGURE 5. Product Frozen Wall Thickness for 8-inch Thermal Maximum Pots

INTERMEDIATE AND LOW LEVEL WASTE TREATMENTMELTER CONDENSATE FROM PHOSPHATE GLASS PROCESSLaboratory Studies - J. E. Mendel

A vacuum distillation procedure for treating sulfate condensate, which was developed at Brookhaven National Laboratory, will be demonstrated in a hot cell with actual sulfate condensate obtained during one of the WSEP phosphate glass runs. In this procedure, a batch distillation of the sulfate condensate (sulfate condensate results from processing sulfate-containing PW-2 feeds by the phosphate glass process) is made at about 20 mm of mercury. Ruthenium is the principal radioisotope in the sulfate condensate and at the low pressure used, it is not appreciably volatilized. A relatively clean separation of nitric acid and sulfuric acid is effected in the batch distillation. The nitric acid can be recycled to a fuels reprocessing plant or redistilled; the sulfuric acid is decontaminated to a sufficient degree that it can be neutralized and asphalted. Ruthenium is recycled to the phosphate glass process with the small amount of phosphoric acid remaining in the pot bottoms.

Sulfate condensate from WSEP Run PG-4 will be used for the in-cell demonstration:

<u>COMPOSITION OF</u>	
<u>PG-4 SULFATE CONDENSATE</u>	
Fe	0.0089 <u>M</u>
Al	0.018
Na	0.017
H ₃ PO ₄	0.097
HNO ₃	0.27
H ₂ SO ₄	0.40

The flowsheet proposed for treatment of this condensate, utilizing a semicontinuous procedure, was tested with simulated solution and tracer ruthenium. The overall decontamination factor was greater than 5×10^3 .

Unique features of the flowsheet were:

- 1) A semicontinuous procedure was used; i.e., four pot volumes were added and boiled off at 60 mm of mercury pressure before the batch boildown was made at 20 mm of mercury pressure.
- 2) Additional phosphoric acid, sufficient to make an M^+/P ratio of about six, was added to prevent precipitation and caking of iron, aluminum, and sodium in the pot bottoms or on the walls. (The metallic ions resulted from entrainment during Run PG-4 and are not necessarily typical of normal sulfate condensate.)

Analyses of the condensate fractions were in agreement with previous findings that most of the ruthenium is volatilized from the pot during the short high temperature period when traces of HNO_3 remain. The test data are summarized below.

Condensate Fraction	Volume, ml	Operating Pressure, mm Hg	BP, °C	Power Input, W	Boiloff Rate, ml/min	Condensate* Activity ^{106}Ru
A	862	60	38-48	84.5	1.25	0.52 $\mu Ci/liter$
B	74	20	36-160	84.5-112	0.7	<0.52
C	7	20	160-250	112-392	<0.5	0.58
D	18	20	250-280	392	1.15	<0.52
E	4	20	280-325	392	1.5	<0.52
Traps	26					<0.52

* Feed activity was $2.55 \times 10^3 \mu Ci/liter$ ^{106}Ru

Tap water was used as coolant for the condensers and was not completely effective in condensing the nitric acid-water mixture. Refrigerated cooling would be required in a production facility.

Four runs were made in the all-glass miniature distillation unit to study the effect of boilup rate upon ruthenium volatility. Ruthenium volatility showed in inverse dependence upon boilup rate; i.e., at higher boilup rates ruthenium evolution was decreased. These results are in agreement with the theory that ruthenium is evolved only after oxidation to RuO_4 . Thus, evolution is kinetically controlled and slower boilup rates allow the oxidation reaction to proceed further toward completion. During these runs it was discovered that 70 to 90 percent of the volatilized ruthenium was not condensed in the tap-water cooled condensers being used and was being carried into the vacuum system. A subsequent run demonstrated that silica gel could be used to trap the noncondensed ruthenium.

Each run consisted of a batch boildown of 250 ml of feed solution of the following composition:

HNO_3	1.0M
H_2SO_4	5.45
H_3PO_4	0.19
Ru	0.0067
^{106}Ru	$\sim 4 \times 10^3 \text{ } \mu\text{Ci/liter}$

The condensate was collected in four increments. Experimental results are shown in Table 6. The ruthenium distribution between condensate increments in Runs SD-18 and 20, made under identical conditions, differed markedly. The difference is thought to be due to irreproducibility of plating and flushing on the condensing surfaces. The conclusion is that very little of the volatilized RuO_4 condenses or is soluble in the

TABLE 6. Sulfate Condensate Distillation: Effects of Boilup Rate

	Run SD-16	Run SD-19	Run SD-17	Run SD-18	Run SD-20
Operating Pressure, mm Hg	20	20	60	60	60
Average Distillation Rate, ml/min	1.0	0.33	1.0	0.33	0.33
Ruthenium Condensate, μCi					
First HNO_3 Fraction	<0.019	<0.018	<0.019	0.016	0.03
Remaining HNO_3 Fraction	<0.007	<0.015	<0.015	0.313	1.02
Transition HNO_3 to H_2SO_4 Fraction	<0.001	<0.003	0.009	0.036	0.24
H_2SO_4 Fraction	<0.013	<0.012	0.042	1.34	0.25
Noncondensable Ruthenium, μCi	not det.	<0.057	1.32	7.45	6.60 ^(b)
Ruthenium in Equipment Flushes, μCi	not det.	<0.12	0.086	1.07	1.40
Ruthenium in Pot Initially, μCi	1070	943	1007	1007	947
Ruthenium in Final Pot Contents, μCi ^(a)	1080	1050	1450	998	919
Material Balance, %	100.7	111.4	144	100.1	98.0
Overall Decontamination Factor	$>1 \times 10^4$	$>4 \times 10^3$	700	100	1000

a. Final pot contents contained solids and were difficult to analyze accurately.

b. Trapped on silica gel bed.

condensate. Most of the ruthenium that reaches the condensate receiver has been reduced to an insoluble form in the condenser and is washed on through by the condensate.

The silica gel bed used in Run SD-20 was 1 1/16 in. in diameter by 9 3/8 in. long and was packed with 6-16 mesh Davison Chemical Corporation silica gel (pretreatment = 1 hr at 300 °C). The bed was held at 60 °C. The first 1 7/8 in. of the bed were completely blackened by deposited ruthenium. At this point, there was a sharp demarcation line and the remainder of the column was colorless. Analysis of the bed showed a sloping front on the loaded zone and indicated that none of the bed was saturated.

Inches Through Bed	Grams Ru per gram SiO ₂
0	4.3×10^{-5}
0.5	2.4×10^{-5}
1.0	1.2×10^{-5}
1.5	0.88×10^{-5}
1.875	0.09×10^{-5}
2.5	0.007×10^{-5}
3.5	$<0.001 \times 10^{-5}$
5	$<0.001 \times 10^{-5}$
7.5	$<0.001 \times 10^{-5}$

Development Studies - W. T. McKean

Cold runs with the Distillation Demonstration Unit (DDU), Figure 6, have been completed using acid mixtures containing 0.19M H₃PO₄ and 0.0067M Ru with sulfuric acid concentrations in the range of 0.5 to 5.45M and nitric acid from 0.455 to 2M. The runs are summarized in Table 7.

The following summarizes the outstanding results:

- 1) Distillation, as a process for cleanup of melter condensate, is a feasible concept.

FIGURE 6. Sulfate Condensate Distillation Demonstration Unit (DDU)

TABLE 7. Summary of Distillation Demonstration Unit

Run No.	Acid Concentration, M		Boilup, gal/hr	Pressure, mm Hg	DF		Comments EDL-102 Cold Test
	HNO ₃	H ₂ SO ₄			HNO ₃	H ₂ SO ₄	
A	1	5.45	2.2	50	855	1559	
F	1	5.45	1.5	25	<3000	1017	
G	1	5.45	1.3	100	73000	1392	
B	1	1	1.7	50	44	5500	
H	1	1	1.9	25	1162	7167	
C	1	0.5	2.1	50	1160	11200	
I	1	0.5	2.2	50	31	309	
N	1	0.5					Analysis not complete
D	0.455	0.5	2.3	50	78	252	
J	0.455	0.5	1.7	30	326	2688	
E	2.0	1.0	1.9	50	76	64	
M	2.0	1.0		25			Not complete

- 2) Decontamination factors for both sulfuric and nitric acid (DF) vary directly with sulfuric acid concentration.
- 3) DF varies inversely with nitric acid concentrations.
- 4) DF varies inversely with distillation pressure. Distillation pressures of 25 mm of mercury resulted in DF values above 300 for the nitric acid stream and 1000 for the sulfuric acid stream.
- 5) In general, distillation with higher concentrations of sulfuric acid permits higher distillation pressures to attain the same DF as is possible at lower sulfuric acid concentration and distillation pressure.
- 6) The results indicate ruthenium may plate out on clean equipment and then be flushed out of the condenser in subsequent runs. As a consequence, the first runs in

the series have higher DF values than later runs. For example, Run C was the first in the series and gave very high DF's while Run I, with identical conditions, was sixth in the series and gave low DF's.

- 7) The high DF values of Run G compared to Run F are unexplained.

It has been shown in other tests by Battelle-Northwest that in laboratory glassware equipment as much as 75 percent of the ruthenium volatilized from the evaporator is noncondensable and is evolved from the system. However no traces were found in attempts to trap ruthenium prior to the DDU vacuum pump and it was concluded that all of the ruthenium stays within the DDU.

Operational problems reported previously with differential pressure cells were resolved by calibrating flow rates in the differential pressure cells prior to each run.

Following completion of the cold runs, some of the operational parts of the DDU equipment were modified to facilitate operation in the hot cell. The DDU equipment and associated vacuum system, feed and transfer lines, and electrical utilities have been installed in C-Cell. Equipment changes for remote operation included:

- 1) Valve orientation and identification on the rack and frame so as to be compatible with manipulator operation.
- 2) Heaters were imbedded in heat-conducting cement and a curing cycle was used to eliminate the last traces of moisture from the cement. This material eliminates air spaces between the heaters and the evaporator wall allowing lower ΔT values and permitting design boilup rates to be reached.

- 3) Installation of a heat exchanger to close the loop on vacuum pump service water. This eliminates the need for retaining effluent vacuum pump service water during hot runs in C-Cell. In order to control the system during hot runs, instrumentation was installed to monitor pH, temperature and liquid level.

During down time for alterations, the tantalum crossover section from the evaporator to the condenser was removed and visually inspected. The interior of the evaporator and differential pressure cell dip legs and the crossover section showed no visual indication of corrosion. Ultrasonic measurements of the evaporator wall indicated no measurable corrosion.

The surface of the Rasching rings in the packed bed on the crossover was coated with a black deposit similar in appearance to the ruthenium deposits which have been reported on the condenser in glassware laboratory runs. The crossover was not dismantled to inspect the demister pad; however, ruthenium appeared to have deposited only in the vertical section of the crossover above the evaporator since the horizontal portion of the crossover and the packed bed of the condenser was clean and had no black deposits. Heavy deposits of ruthenium were found on the interior part of the asbestos gasket exposed to the vapor at the flange between the evaporator and the crossover. No deposits were found at the flange between the condenser and the crossover.

A threaded stainless steel fitting on the liquid sampler at the base of the condenser was found to have corrosion on the threads. Ultrasonic measurements of acid receiving tanks will be made to ascertain if significant corrosion of the stainless steel had taken place.

Water boilup tests and three cold runs have been made in C-Cell. The feed composition of the cold runs was identical to the out-of-cell cold Runs D and J listed in Table 7. These runs were duplicates of cold Runs D and J and were made to determine how closely DF values can be reproduced and to find if there is a trend toward lower DF values as subsequent runs are made. Such a trend would indicate ruthenium retention and carry over from early runs to later runs. Sample analysis for these runs is in progress.

Maximum average boilup rate for these runs was at the design capacity of 2.5 to 3.0 gal/hr. During a water evaporation boiloff, heater surface-to-water temperature ΔT was 50 to 70 °F and during evaporation of sulfuric acid, boilup rates were 0.25 to 0.60 gal/hr with a ΔT of about 250 °F.

During the down time for modification, a sludge layer about 1/4 to 1/2 in. thick was found on the bottom of the evaporator vessel. The layer was compact, though it could be scraped away in small pieces with some difficulty. It was insoluble in various combinations of nitric, hydrochloric, and sulfuric acids, but was readily dissolved in 1M sodium hydroxide. Sludge accumulation is suspected in the present runs since the two banks of heaters on the bottom of the evaporator have had increasing surface temperatures for each subsequent run.

Occasional tests of the vacuum system indicate no deterioration of the system or development of leaks. The minimum operational pressure which can be controlled is 20 mm of mercury, a value which has remained the same throughout the run period. Occasional leak tests are made by evacuating the system and isolating it at 20 mm of mercury overnight. After 12 hr the normal leak rate raises the pressure to about 100 mm of mercury.

EVALUATION OF SOLIDIFIED WASTE PRODUCTS

NONRADIOACTIVE LABORATORY STUDIES

Study of High Temperature Storage of Solidified Waste Products Containing Sulfate - D. W. Brown and J. E. Mendel

A laboratory investigation of molten core storage was started using simulated spray solidifier PW-2 product. This product contains 17 wt% SO_4 and is considered to pose the most severe problems for molten core storage. Two principal effects are being studied: 1) the possibility of the melt developing nonhomogeneities which can produce hot spots, and 2) the possibility of excessive pressure buildup due to SO_3 evolution.

The release of SO_3 from simulated spray solidifier PW-2 product was measured as a function of time over the temperature range 550 to 1000 °C. The release of SO_3 was followed gravimetrically, assuming all weight lost was SO_3 . The results, given in Figure 7, show an appreciable rate of sulfate decomposition even below the melting point of the product (~725 °C).

Products were examined after being held 120 days at 550, 650, and 750 °C. Separation into two distinct phases occurred at 750 °C. The upper phase (60 vol%) was a dark-green glass, the lower phase was a pale green microcrystalline material. X-ray fluorescence measurements indicated over 90 percent of the rare earths and 70 percent of the zirconium was the lower phase. The radioactive isotopes of these elements are major heat producers and segregation of this magnitude in a molten core would definitely produce a hot spot.

*FIGURE 7. Sulfate Loss from
Spray Solidifier PW-2
Product Held in Air*

Evidence that a similar phase separation was occurring, but at a much slower rate, could be seen in the sample held at 650 °C. The sample held at 550 °C was homogeneous.

The evolution of SO_3 during molten core storage of spray solidifier PW-2 product is being studied in a small platinum-lined Hastelloy B pressure vessel. The vessel contained a 150 g cylinder of simulated spray solidifier PW-2 product 5/8 in. in diameter by 10 in. long. The bottom 6 in. of the melt is being held at 800 °C. The temperature of the remaining melt is reduced to 375 °C at the top to simulate the temperature profile of an air-cooled storage container with a molten core. The temperature in the 2 in. vapor space above the product is 300 °C.

The vessel was evacuated prior to sealing and heating. It has now been held at temperature for 6 weeks and no increase in pressure has occurred. Indications are that sulfate decomposition in the spray solidifier PW-2 melt is reversible. At the end of the experiment, the melt will be sectioned and analyzed to determine if sulfate has migrated from the hot end to cooler portions of the melt.

Product Characterization - Continuation of Laboratory
Evaluation of PW-2 Phosphate Glass Samples - J. H. Kleinpeter

Reference has been made in previous reports* to a series of tests which are being made to characterize the PW-2 phosphate glass from the Brookhaven phosphate glass process and to determine the effect of thermal history on the physical properties. The chemical makeup of the melt is standard PW-2 composition with 3.13M H_3PO_4 added to give an M^+/P of 1.0. The thermal parameters chosen for study and the values used were:

1. Melt temperature, 1000 °C, 1200 °C.
2. Hours at melt temperature, 1 hr, 25 hr.
3. Cooling rate °C/hr from 900, 300, 100 °C.
4. Storage temperature for eight weeks, 100, 600 °C.

Sixteen samples were prepared using every possible combination of these values. The samples were mounted and polished and photomicrographs were made. Those samples which had been stored at the high temperature were by far the most crystalline, but all samples exhibited some crystallinity.

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* BNWL-889, 68-2, and BNWL-923, 68-3

Thermal Conductivity

Comparative measurement of thermal conductivity was not satisfactory because there was a low-temperature melting fraction oozing to the surface of the sample during the test. This material wetted the granular insulation around the specimen and left voids in the sample.

Thermal conductivity is a function of thermal diffusivity, heat capacity and density. This relationship is expressed by:

$$k = \alpha \rho C_p,$$

where,

k = thermal conductivity, cal/(sec) (cm) (°C)

α = thermal diffusivity, cm²/sec

ρ = density, g/cm³

C_p = heat capacity, cal/(g) (°C).

Thermal Diffusivity

Thermal diffusivity was measured on all sixteen samples at several temperatures between 30 and 400 °C. Values are reported in Table 8. In general, the samples seem to fall into two types. One with a low diffusivity which changes only slightly with temperature, and the other with a somewhat higher value which decreases sharply with increasing temperatures. Most of the more crystalline samples fall into the latter group.

Heat Capacity

The heat capacity of three samples has been determined using a Perkin-Elmer differential Scanning Calorimeter. Of the three samples, one was from the group having a high temperature sensitive thermal diffusivity and the other two were from the low, less temperature-sensitive group. The heat capacity and the thermal conductivity of these three samples are recorded in Table 8.

PRODUCT MEASUREMENTS, TESTING, AND STORAGESolids Storage Engineering Test Facility (SSETF) - R. J. Thompson

The first radioactive WSEP waste pot was assembled into an environmental test pod in SSETF to begin the interim storage studies. The first pot is from the phosphate glass Run PG-4. The pot material is mild steel and contains PW-2 solidified waste. The waste fission product heat is relatively low (2.2 kW at the time of installation) and will be augmented by pod electrical heat to achieve desired air storage temperatures.

Product Characterization

Several pot calorimetry values were obtained in B-Cell.

1. PG-6   9.5 kW
2. SS-5   5.3 kW
3. SS-5   8.3 kW
4. SS-6   11.1 kW
5. SS-7   12.7 kW

All heat values are related to the run date.

TABLE 8. Characterization of PW-2 Phosphate Glass Samples

Sample	A	B	C	D	$10^{-4} \alpha$, cm ² /sec	Temp, °C	C_p , cal/ (g) (°C)	Temp, °C	$10^{-3} k$, cal/(sec) (cm) (°C)	Temp, °C	ρ , g/ cm ³
2	1200	1	100	600	48	34					2.92
					47	34					
					48	34					
					52	55					
					51	90					
					41	126					
					39	205					
					37	254					
					33	325					
					37	400					
3	1200	25	100	600	46	30					2.89
					47	30					
					46	75					
					40	124					
					35	200					
					30	250					
					36	265					
					33	325					
					33	400					
4	1000	1	30	100	37	29	0.192	50-100	1.97	75	2.94
					35	75	0.202	100-150	2.02	125	
					33	126	0.199	150-200	1.87	175	
					31	206	0.220	200-250	2.00	225	
					31	252	0.234	250-300	2.06	275	
					31	325	0.277	300-350	2.44	335	
					29	355	0.306	350-400	2.61	375	
					28	400					
5	1000	1	100	100	30	30					
					30	31					
					30	75					
					29	125					
					29	200					
					28	250					
					28	325					
6	1000	25	330	100	43	31					2.81
					39	75					
					37	135					
					36	202					
					35	223					
					34	255					
					35	325					
					34	400					
7	1000	1	30	600	36	31					2.99
					37	32					
					37	32					
					37	50					
					35	77					
					34	125					
					34	220					
					29	250					
					29	276					
8	1000	1	100	600	42	32					2.98
					42	58					
					35	75					
					38	82					
					33	208					
					28	250					
					30	325					
					26	400					
					27	400					
9	1200	25	25	600	46	34					2.93
					40	37					
					40	77					
					36	81					
					36	125					
					32	185					
					28	250					
					31	270					
					30	325					
					27	399					

TABLE 8. Characterization of PW-2 Phosphate Glass Samples (contd)

Sample	A	B	C	D	$10^{-4} \alpha, \text{cm}^2/\text{sec}$	Temp, °C	$C_p, \text{cal}/(\text{g})(^\circ\text{C})$	Temp, °C	$10^{-3} k, \text{cal}/(\text{sec})(\text{cm})(^\circ\text{C})$	Temp, °C	$\rho, \text{g}/\text{cm}^3$
10	1200	1	30	600	44	28	0.162	50-100	1.96	75	2.88
					44	29	0.166	100-150	1.91	125	
					42	75	0.171	150-200	1.91	175	
					39	126	0.199	200-250	2.06	225	
					37	200	0.212	250-300	2.13	275	
					35	254	0.226	300-350	2.15	325	
					33	325	0.282	350-400	2.59	375	
					33	335					
					33	400					
11	1000	25	100	100	28	32					2.89
					28	59					
					28	75					
					27	131					
					27	200					
					23	255					
					28	266					
					29	321					
					32	347					
					25	362					
					26	400					
12					29	32	0.181	50-100	1.53	75	2.91
					27	35	0.181	100-150	1.48	125	
					29	57	0.204	150-200	1.66	175	
					29	75	0.222	200-250	1.74	225	
					27	200	0.210	250-300	1.65	275	
					27	215	0.251	300-350	1.97	325	
					27	250	0.257	350-400	1.94	374	
					27	321					
					26	400					
13	1200	25	30	100	35	33					2.90
					31	52					
					31	75					
					31	77					
					35	112					
					30	159					
					34	165					
					34	192					
					36	200					
					34	219					
					38	325					
					34	397					
					34	400					
14	1200	1	100	100	29	28					2.92
					29	30					
					26	123					
					21	213					
					26	250					
					26	235					
					25	352					
					25	400					
15	1200	25	30	600	30	34					2.92
					30	35					
					31	85					
					30	92					
					29	97					
					29	125					
					29	206					
					28	252					
					28	325					
16	1000	25	100	600	49	28					2.93
					44	80					
					41	125					
					37	208					
					35	250					
					34	325					
					32	390					
					29	400					
17	1200	25	100	100	29	32					2.91
					27	86					
					26	125					
					26	144					
					26	204					
					26	250					
					26	257					
					23	325					
					25	400					

Note:

A = Melt Temperature.

B = Hours in Melt.

C = Cooling Rate °C/hr.

D = Eight Week Storage Temperature.

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