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QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
FIXATION OF RADIOACTIVE RESIDUES
AUGUST, SEPTEMBER AND OCTOBER 1969

December 1969

AEC RESEARCH &
DEVELOPMENT REPORT

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AUGUST, SEPTEMBER AND OCTOBER, 1969

Edited by

A. G. Blasewitz

Chemical Technology Department
Chemistry and Metallurgy Division

December, 1969

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* *Reference notation used in this report.*

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	<u>1969</u>		
First		BNWL-1074	69-1
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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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QUARTERLY PROGRESS REPORT
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FIXATION OF RADIOACTIVE RESIDUES

August, September, and October, 1969

INTRODUCTION

This progress report is the forty-third 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

To date, 26 radioactive demonstrations in WSEP have been completed with simulated wastes containing approximately 38 MCi of mixed radionuclides being converted to solids.

During the report period, approximately 6.2 MCi of radioactivity were **successfully** processed in three phosphate glass solidification runs in WSEP. A simulated Purex Waste 4m (PW-4m) was processed during all three runs and was the first radioactive demonstration of the phosphate glass process with this waste (a low salt, high fission product waste resulting from the reprocessing of a power reactor fuel with an exposure of 45,000 MWd/tonne at 30 MW/tonne). In the final run, a heat rate density of 315 W/liter was attained in the product. This is equivalent to that which would result from solidifying waste from the reprocessing of power reactor fuel aged 0.25 years.

A simulated PW-4m waste was solidified during a developmental spray solidifier run in which the calcined waste was fluxed to a semivitreous solid in the waste storage container. In another developmental run, a simulated fast reactor waste (a low salt, high fission product containing waste resulting from the reprocessing of a fast reactor fuel with an exposure of 100,000 MWd/tonne at 200 MW/tonne) was solidified, and the process scheme appears suitable for demonstration with the WSEP spray solidifier.

Gas sampling techniques that have been initiated will permit better definition of the type, form, and extent of radioactivity in the process off-gas from the solidifiers.

Environmental testing of waste containers in the Solids Storage Engineering Test Facility was initiated with testing of waste containers from WSEP Runs SS-4 and PG-6.

OVERALL STATUS OF WSEP RADIOACTIVE DEMONSTRATIONS

K. J. Schneider

Table 1 shows the overall status of the radioactive demonstrations in WSEP as of October 31, 1969. A total of 26 demonstrations have been completed: 6 in the pot calcination process, 10 in the phosphate glass solidification process, and 10 with the spray solidification process. Approximately 38 MCi of mixed radionuclides representing waste from about 253,000 MWd of electrical power have been converted to solids. This amount of waste represents 82 days of all the nuclear power capacity in the United States as of September 1, 1969. The total on-line processing time for solidification of these wastes was 60 days.

TABLE 1. Overall Status of WSEP Radioactive Demonstrations

	solidification Methods			Total
	Pot	Spray	Phosphate Glass	
Runs Completed	6	10	10	26
Megacuries Solidified	4.0	17.5	16.6	38
Equivalent Tonnes(a)				
Processed	11.3	14.6	12.0	38
MWd _e Represented by Waste(b)	75,000	98,000	80,000	253,000
Tonne/Day Rate(a)	0.6 to 1.0	0.5 to 0.9	0.3 to 0.7	--
Maximum kW in One Pot	5.1	12.7	8.5(c)(d)	140(f)
Maximum W/R in Pot	85	205	315(d)(e)	--
Maximum Centerline Temperature in Pot, °C	940	930	780	--
Liters Solid/Tonne(a)	40 to 50	30 to 65	50 to 100	--

(a) Tonne is a metric tonne (1000 kg, or 2205 lb) of original fuel.

(b) Assuming 33% thermal efficiency and 20,000 MWd/tonne.

(c) True maximum has not yet been demonstrated for 8-in. diameter pots where the calculated maximum is 11.7 kW.

(d) This value is for 6-in. diameter pots, whereas the comparable values for the pot and spray solidifiers are for 8-in. diameter pots.

(e) True maximum has not yet been demonstrated for 8-in. diameter pots where the calculated maximum is 195 W/l.

(f) Total kilowatts encapsulated to date.

PHOSPHATE GLASS SOLIDIFICATION

LABORATORY STUDIES

Corrosion Studies - R. F. Maness

A laboratory-scale PW-4m phosphate glass run was made with feed solution containing 0.05M fluoride. The run was made at 1100 °C with semicontinuous feed addition to an alumina crucible equipped with an overflow weir. Total time at temperature was 72 hr, while feed was added semicontinuously for 27 hr. Platinum specimens exposed to the melt and to the melt-vapor interface corroded at a rate of 0.0015 in./year.

A specimen exposed to the vapor phase corroded at a rate of 0.00013 in./year. No evidence of embrittlement or cracking was noted in any case. A Nionel specimen exposed in the off- as line corroded at a rate of 0.012 in./year.

RADIOACTIVE DEMONSTRATIONS - J. L. McElroy, J. N. Hartley,
B. W. Bentley, and 7. Drager*

Phosphate Glass Solidification Run PG-8

During Run PG-8, simulated PW-4m waste equivalent to 0.96 tonnes of power reactor fuel (45,000 MWd/tonne at 30 MW/tonne) after 1.4 years out of the reactor was solidified to yield 195 kg of phosphate glass product. The 66.7 liters of radioactive product produced a self-generating heat rate density of 132 W/liter (8470 W total by pot calorimetry) in an 8-in. diameter mild steel receiving pot. This was the first phosphate glass run with the high-burnup, 45,000 MWd/tonne waste. Table 2 lists the PW-4m feed composition for Runs PG-8, PG-9, and PG-10.

The denitrator-evaporator was fed a total of 660 liters of PW-4m feed (622 liters/tonne) in 39.5 hr at an average feed rate of 13.5 liters/hr. The feed contained approximately 2,300,000 Ci of radioactivity including 9800 Ci of radio-ruthenium. Foaming problems in the denitrator were avoided by the addition of a commercial silicone antifoam agent as in previous runs.** Operating parameters for Runs PG-8, PG-9, and PG-10 are summarized in Table 3. Also included are data for Run PG-7 which was discussed in the previous quarterly.***

* On assignment from Brookhaven National Laboratory.

** BNWL-834, 68-1

*** BNWL-1168, 69-2

TABLE 2. PW-4m Feed Composition for Runs PG-8, PG-9, and PG-10

Component	PG-8	PG-9	PG-10
H ⁺	--	--	--
Fe ⁺³	0.05M ^(a)	0.05M ^(a)	0.064M ^(a)
Cr ⁺³	0.012	0.012	0.012
Ni ⁺²	0.008	0.008	0.008
Al ⁺³	0.001	0.001	0.12
Na ⁺	0.10	0.10	0.10
PO ₄ ⁻³	0.003	0.003	0.003
NO ₃ ⁻	~6.5	~6.5	~6.5
SO ₃ ⁻²	~0.01	~0.01	~0.01
MoO ₄ ⁻² + Tc as MoO ₄ ⁻²	0.16	0.16	0.16
Sr ⁺²	0.036	0.036	0.036
Ba ⁺²	0.041	0.041	0.041
Rb ⁺ + Cs ⁺ (added K ⁺)	0.092	0.092	0.092
(Y + Re) ⁺³	0.274	0.274	0.53
ZrO ⁺²	0.143	0.143	0.143
Ru ⁺⁴	0.0032	---	0.01
Ru ⁺⁴ (added Fe ⁺³)	0.078	0.080	0.072
Rh ⁺² (added Co ⁺²)	0.013	0.013	0.0133
Pd ⁺² (added Ni ⁺²)	0.042	0.042	0.0425
Ag ⁺² (added Cu ⁺²)	0.0016	0.0016	0.0016
Cd ⁺² (added Cu ⁺²)	0.0025	0.0025	0.0025
TeO ₄ ⁻³ (added SO ₄ ⁻³)	0.014	0.014	0.014
<u>Additive to Feed</u>			
Na ⁺	1.40	1.40	1.40
Fe ⁺³ (b)	0.05	--	--
Al ⁺³ (b)	0.40	0.45	0.45
PO ₄ ⁻³	5.07	5.07	5.25

(a) Molarities are at 378 liters/tonne.

(b) Nominal Fe additive is 0.45M, however excess Al in the waste from the processing plant is used as a substitute for Fe.

TABLE 3. *Summary Data for WSEP Phosphate Glass Runs PG-7, PG-8, PG-9 and PG-10*

Operational Data	PG-7	PG-8	PG-9 ^(a)	PG-10 ^(a)
Time of Run	7/7/69- 7/10/69	8/11/69- 8/13/69	9/22/69- 9/23/69	9/24/69- 9/25/69
Operating Mode	A	88% A - 12% B	A	A
Feed Type	PW-2	PW-4m	PW-4m	PW-4m
Feed, liters/tonne	644	622	1023	1198
Pot Diameter, in.	8	8	6	6
Pot Material	310 SS	Mild Steel	304L SS	304L SS
Total Feed to Denitrator-Evaporator, liters	806	660	530	580
Total Feed to Melter, liters	675	610	423	420
Total Feed Time, hr	44.2	49.5	22	24
Average Feed Rate, liters/hr	18.2	13.3	24	24.2
Equivalent Waste to Solidifier, tonne	1.25	1.1	0.52	0.48
Equivalent Waste to Receiver pot, tonne	1.05	0.98	0.41	0.35
Volume Reduction, feed volume/product volume	9.5	9.1	15.1	14
Volume Reduction if Feed were at 378 liters/tonne	5.6	5.6	5.6	4.4 ^(d)
Filled Pot Data				
Total Curies in Feed to Pot ^(b)	2,300,00	2,850,000	1,200,000	2,700,000
Total Curies of Ru in Feed to Solidifier ^(b)	9800	13,000	9100	
Product in Pot, liters	67	71	28	30
Product in Pot, liters/tonne	83	67.6	68	85.7
Heat Generation Rate in Pot, W ^(c)	8500	9800	4900	9500
Product Heat Rate Density, W/liter	127	138	175	315
Equivalent Aging Time, years	1.4	0.46	0.95	0.25
☐ Temperature in Warm Furnace, °C	827	853	NA	NA
☐ Temperature in Air Cooled Furnace	710	782	565	827
☐ Temperature in Air, °C	659	749	NA	776
☐ Temperature in Water, °C	475	548	290	553
Effective Thermal Conductivity of Product in Warm Furnace, W/(m ²)(°C/m)	1.52	1.78	NA	NA
Effective Thermal Conductivity of Product in Air Cooled Furnace, W/m ² (°C/m)	0.95	0.9	1.21	1.20
Effective Thermal Conductivity of Product in Air, W/(m ²)(°C/m)	0.94	0.86	NA	1.12
Effective Thermal Conductivity of Product in Water, W/(m ²)(°C/m)	0.77	NA	0.98	0.87

(a) Preliminary data

(b) By feed analysis

(c) By pot calorimetry data

(d) Volume reduction is low because rare earths were in excess in the PW-4m feed. (0.53M actual versus 0.27M nominal)

NA Not available

The bulk density of the glassy product in the receiver pot was 2.9 kg/liter. The self-generating heat rate density of 132 W/liter produced a pot centerline temperature of 660 °C with the pot hanging in air to give an effective thermal conductivity of $0.94 \text{ W/(m}^2)(^{\circ}\text{C/m)}$ [$0.54 \text{ Btu/(hr)(ft}^2)(^{\circ}\text{F/ft)}$]. The effective thermal conductivity data for the phosphate glass product is highly dependent on storage conditions as shown in Table 4. The significant increase in thermal conductivity at the higher storage temperature is the result of the softening and melting of the product at approximately 700 °C.

TABLE 4. PG-8 Receiver Pot Temperatures and Product Thermal Conductivity Data

Receiver Pot Location	Temperature, °C			$k_{\text{eff}} \text{ W/(m}^2)(^{\circ}\text{C/m)}$
	Centerline	Wall	AT	
Warm Furnace	820	600	220	1.52
Air Cooled				
Furnace	710	360	350	0.95
Air	660	300	360	0.94
Water	475	42	433	0.77

The melter furnace was operated at an average temperature of 1230 °C, while the internal melt control temperature for controlling feeding of the melter was 970 °C and the melt pool near the bottom of the melter was 1040 °C. Better interpretation of internal melter operating conditions and less reliance on visual access for trouble-free melter operation during melter feeding was realized by continuously monitoring the melt temperature at a point 1/2 in. below the weir overflow. This temperature had previously been recorded on a slower responding multipoint recorder. Melt was continuously discharged from the melter to the pot through the weir overflow at a rate of 1.6 liters/hr. The volume reduction obtained was 9.2 liters of adjusted feed to the denitrator per liter of glass produced (5.6 if the feed concentration was 378 liters/tonne).

The denitrator agitator began vibrating early in the run and was not used during the last three-fourths of the run; however, no obvious detrimental effects were noted. A new scheme was initiated during PG-8 to prevent plugging of the liquid level and specific gravity dip tubes. The method involved venting the tubes to the denitrator vapor space approximately four times each hour for a period of 5 to 10 min. This allowed the denitrator liquid to rise in the tubes and effectively wash them. The method worked well during PG-8.

The final 12% of the run was conducted under Mode B operation (concentration of PG-9 waste during PG-8) by feeding Purex high level waste from the evaporator feed tank (TK-112) to the waste evaporator (TK-113). The Mode A and B arrangement for Runs PG-7 through PG-10 is shown in Figure 1. The radioactivity distribution during each mode of operation and for the total run is presented in Table 5. During Run PG-8, 0.62% of the ruthenium and 0.03% of the nonvolatiles (represented by CePr-144) that were fed to the denitrator were volatilized and/or entrained from the denitrator and accumulated in the auxiliary evaporator. A total of 4.0% of the ruthenium and 0.04% of the nonvolatiles in the feed to denitrator were volatilized and/or entrained from the melter.

The fraction of feed radioactivity that accumulated in the acid fractionator for ruthenium was 4.3×10^{-4} during Mode A operation and 7.5×10^{-3} during Mode B. For the entire run, the fraction was 1.4×10^{-3} for ruthenium and 4.2×10^{-8} for nonvolatiles.

FIGURE 1. Mode A and B Arrangement for PG-7, PG-8, PG-9 and PG-10

TABLE 5. *Radioactivity Distribution for Runs PG-7, PG-8, PG-9 and PG-10*

	FRACTION OF FEED RADIOACTIVITY									
	PG-7		PG-8						PG-9	
	MODE A		MODE A		MODE B		TOTAL RUN			
	Ru-106	CePr-144	Ru-106	CePr-144	Ru-106	CePr-144	Ru-106	CePr-144	Ru-106	CePr-144
Melter Condensate (TK-117)	1.4×10^{-1}	3.5×10^{-4}	3.2×10^{-2}	3.4×10^{-4}	9.4×10^{-2}	8.3×10^{-4}	4×10^{-2}	4×10^{-4}	4×10^{-2}	5×10^{-4}
Evaporator Bottoms (TK-113)	8.0×10^{-3}	4.1×10^{-3}	6.2×10^{-3}	2.9×10^{-4}	3.2×10^{-3}	1.6×10^{-4}	6.2×10^{-3}	2.9×10^{-4}	7.5×10^{-3}	5×10^{-4}
Fractionator Bottoms (TK-115)	1.6×10^{-3}	1.5×10^{-6}	4.3×10^{-4}	NA	7.5×10^{-3}	4.2×10^{-6}	1.4×10^{-3}	4.2×10^{-8}	1.5×10^{-4}	NA
Fractionator Condensate (TK-116)	1.0×10^{-6}	2.8×10^{-8}	3.1×10^{-6}	NA	7.8×10^{-7}	1.0×10^{-9}	2.8×10^{-6}	3.7×10^{-10}	6×10^{-7}	2×10^{-8}
Offgas to Scrubber	6.5×10^{-9}	4.40×10^{-7}	3.2×10^{-6}	2.6×10^{-10}	6.6×10^{-5}	6.5×10^{-9}	1.3×10^{-5}	1.6×10^{-7}	NA	NA
Offgas from Scrubber	9.6×10^{-8}	1.5×10^{-10}	---	---	---	---	9.9×10^{-9}	2.7×10^{-11}	NA	NA
Offgas to Stack	$<1 \times 10^{-12}$	4×10^{-12}	---	---	---	---	$<2.6 \times 10^{-10}$	4.8×10^{-12}	NA	NA
Ratio (fract. cond) Activity to 10CFR20	2.5×10^3	55	5.2×10^3	54	4.5×10^4	2.4×10^2	5.6×10^3	89		
Ratio (Final Offgas) Activity to 10CFR20	4.5×10^2	7.2×10^2	---	---	---	---	3.3×10^2	1.1×10^2	NA	NA
Ratio (Offgas to Stack) Activity to 10CFR20	$<4.0 \times 10^{-4}$	$<5.8 \times 10^{-6}$	---	---	---	---	$<1.8 \times 10^{-4}$	2.8×10^{-4}	NA	NA

The cumulative decontamination factor (DF) for ruthenium across the auxiliary evaporator was 420. Instantaneous decontamination factors (DF's) averaged about 10^3 during Mode A and Mode B. The instantaneous DF's across the evaporator for nonvolatiles represented by CePr-144 averaged about 2×10^3 during Mode A and 4×10^5 during Mode B.

The cumulative DF across the fractionator was 590 for ruthenium. The instantaneous DF's for ruthenium averaged 100 during Mode A and 360 during Mode B. The instantaneous DF's for CePr-144 averaged less than 10 during both Mode A and Mode B.

The accumulated fractionator condensate contained a fraction of feed radioactivity equal to 2.8×10^{-6} for ruthenium and 3.7×10^{-10} for CePr-144. These values were above 10CFR20 release limits by factors of 5.6×10^3 and 89, respectively.

The average **concentration** of radioactivity in the final process off-gas leaving the scrubber was $6.5 \times 10^{-8} \mu\text{Ci}/\text{cm}^3$ for ruthenium and 4.4×10^{-8} for CePr-144. These values were above 10CFR20 release limits by factors of 330 and 110, respectively. However, before the gas is released to the environment, the final process off-gases are combined, filtered, and significantly diluted (about 3×10^3 times) by the building ventilation before finally entering a 150 ft stack for release to the atmosphere. The ratio of activity leaving the stack to 10CFR20 release limits was less than 1.8×10^{-4} for ruthenium and 2.8×10^{-4} for CePr-144: well below release limits.

Phosphate Glass Solidification Run PG-9

During Run PG-9, a simulated waste equivalent to that resulting from the reprocessing of 0.52 tonne of power reactor fuel (45,000 MWd/tonne at 30 MW/tonne) with an equivalent aging time of 0.95 years was reduced to 28 liters of phosphate glass product. The product, which was collected in a 6 in. diameter pot, produced a self-generating heat rate density of 175 W/liter (4900 W total based on pot calorimetry data).

The denitrator was fed 530 liters of adjusted PW-4m type feed (1023 liters/tonne) in 22 hr at an average rate of 24 liters/hr. (See Table 2 for feed composition.) The feed contained approximately 1,200,000 Ci of radioactivity including 9100 Ci of radoruthenium. **As** in most previous runs* the continuous addition of a commercial silicone antifoam agent to the denitrator prevented all signs of foaming.

Foaming in the melter was generally mild, but on one occasion early in the run the melt level became high. This probably resulted from maintaining an excessive feed rate from the denitrator airlift to the melter for a short time (1/2 to 1 hr). The excessive feed rate caused the melt to swell and foam, and as the cool surface of the foaming melt rose up over the internal weir, the overflowing melt within the weir became solidified. This prevented additional melt from flowing to the receiver. The weir is normally thawed by terminating melter feeding until the melter temperature recovers sufficiently to thaw the frozen melt within the weir. On this occasion, the cooling air on the external freeze drain valve was also decreased to gain additional heat on the external portion of the weir. As a result, the freeze-drain valve thawed and the

* BNWL-834, 68-1

melter was inadvertently drained to the receiver pot. After the freeze-drain valve was resealed the melter was refilled to its normal operating volume of approximately 5 liters, and the run was completed by the weir overflow method of filling the receiver pot. (At the end of the run the melter was drained by thawing the freeze-drain valve.) The rate of discharge of radioactive glass from the melter to the receiver pot was approximately 1.4 liters/hr.

The 6-in. diameter 304L stainless steel receiver pot was maintained at 600 °C and was uniformly filled without the entering melt striking the pot wall or thermowells. All previous melt receivers used in WSEP were either 8 or 12 in. in diameter. A 6-in. diameter receiver pot allows processing shorter cooled wastes with a higher heat rate density than is possible with larger diameter pots. In this respect, the 175 W/liter heat rate density in Run PG-9 was a successful trial run for Run PG-10 where the internal heat rate density was increased to 315 W/liter.

During Run PG-9, 0.75% of the ruthenium and 0.05% of the CePr-144 (representing nonvolatiles that were fed to the denitrator) were volatilized and/or entrained from the denitrator and accumulated in the auxiliary evaporator. A total of 4% of the ruthenium and 0.05% of the CePr-144 in the feed to the denitrator were volatilized and/or entrained from the melter.

The fraction of feed radioactivity that accumulated in the acid fractionator was 1.5×10^{-4} for ruthenium. The accumulated fractionator condensate contained a fraction of feed radioactivity equal to 6×10^{-7} for ruthenium and 2×10^{-8} for nonvolatiles. These values were above 10CFR20 release limits by factors of 1900 and 400, respectively.

Phosphate Glass Solidification Run PG-10

Phosphate glass Run PG-10 was made immediately following Run PG-9. Both runs were completed in a time period of 75 hr, even though a feed pump required remote changeout between runs. This was the first time that two consecutive runs had been attempted.

During Run PG-10, simulated waste equivalent to 0.35 tonnes of power reactor fuel (45,000 MWd/tonne at 30 MW/tonne) with an equivalent aging time of 0.25 years was solidified to produce 30 liters of phosphate glass product with an internal heat rate density of 315 W/liter (9500 W total based on pot calorimetry data). This heat rate density was 35% greater than that produced in any previous WSEP product and 45% greater than that for the "hottest" phosphate glass product which was produced during Run PG-9. No difficulties were encountered in filling the 6-in. receiver or in handling the pot after it was filled. With the receiver setting in the air-cooled furnace at the end of the run, the centerline temperature was 827 °C and the centerline-to-wall temperature difference was 378 °C. For this condition of a partially molten product (remelt temperature of approximately 700 °C), the effective thermal conductivity was 1.20 W/(m²) (°C/m) [0.69 Btu/(hr) (ft²) (°F/ft)]. When the receiver was placed in water storage, the centerline temperature decreased to 553 °C. In water the centerline-to-wall temperature difference of 520 °C gave an effective thermal conductivity for the more solid product of 0.87 W/(m²) (°C/m) [0.5 Btu/(hr) (ft²) (°F/ft)].

The denitrator was fed approximately 580 liters of adjusted PW-4m type feed (1198 liters/tonne) in 24 hr at an average feed rate of 24.2 liters/hr. Approximately 160 liters of the

feed remained in the denitrator at the end of the run. All of the 2,700,000 Ci of radioactivity in the feed to the denitrator was obtained from a radiocerium solution. Since no Purex 1WW was used in the feed makeup, no antifoam agent was needed for foam suppression in the denitrator evaporator. (Purex 1WW contains small amounts of dibutyl phosphate which causes excessive foaming in the denitrator.)

Some plugging difficulties were encountered with the denitrator dip tubes and airlift system when the denitrator was initially concentrated to a 125 °C boiling point. After the temperature was decreased to 122 °C and following some flushing, there were no further plugging difficulties. Since the PG-10 feed contained an excess of rare earths (0.53M versus 0.27M desired), it is believed that this was responsible for the change in the characteristics of the denitrator-evaporator concentrate as both PG-8 and PG-9 were successfully operated at 125 °C with nominal PW-4m feed.

The dip tube flushing scheme that was initiated during PG-8 gave highly satisfactory results during both PG-9 and PG-10.

With the melter operating at a furnace temperature of 1250 °C, the average melt pool temperature was nearly 1200 °C (the melt temperature actually equalled the furnace temperature when no feed was entering the melter), and the melt rate to the receiver was approximately 1.4 liters/hr. The melt was overflowed through the unheated melter weir until the end of the run when it was batch-dumped through the freeze valve to completely empty the melter. Receiver temperatures were controlled at approximately 600 °C to prevent possible stalagmite formation.

Because the heat rate density of the PG-10 waste was considerably higher than that for PG-9, it was necessary to change feed batches between runs and to also flush the PG-9 concentrate from the denitrator. Removal of the PG-9 receiver pot and installation of the PG-10 receiver required only 2.5 hr. The feed pump changeout was made between runs as there were definite indications, both before and after PG-9, that the pump was failing. The indications included a high starting amperage and a fluctuating amperage during operation. The pump had been used for three complete runs.

SPRAY SOLIDIFICATION

LABORATORY STUDIES

Corrosion Studies - R. F. Maness

Corrosion of 304L stainless steel was evaluated at 950 °C in melts formed from PW-4m calcine containing calcium and zinc oxides plus: 1) borosilicate glass frit, 2) boron oxide, 3) sodium metaphosphate, 4) lead-bearing glass frit, and 5) high alkali glass frit. Corrosion rates of 304L specimens exposed to the melt-vapor interface were about 0.12 in./year in the melts formed with borosilicate glass frit and with boron oxide. The other systems were more corrosive to 304L with corrosion rates equal to approximately 1.0 in./year. Corrosion rates of 304L interface specimens at 850 °C in melts formed with borosilicate glass frit, sodium metaphosphate, and high alkali glass frit were 0.008, 0.6, and 2.5 in./year, respectively. In all cases, specimens exposed to the melt-vapor interface corroded at a higher rate than those exposed to the melt phase only.

ENGINEERING STUDIESIn-Pot Melting Studies - J. D. Moore

Developmental spray solidifier Run DSC-35 was carried out to test an operable process for spray solidifying PW-4m liquid waste using the product storage pot as the melting vessel (in-pot melting). Simulated PW-4m with added Ca(OH)_2 and ZnO (see Table 6) was calcined in the developmental spray calciner during an 18-hr run. Powdered B_2O_3 flux was added to the 304L stainless steel melt storage pot which was heated to 950 °C. A summary of the operating conditions and results is given in Table 7.

Feed flow stability at an average feed rate of 17.9 liters/hr was adequate even though the feed contained about 70 vol% solids. Although no problems were encountered with feed flow in this run, agitation of the feed must be maintained to prevent solids from settling and clogging the feed lines and feed control valve. For this reason, it would be desirable to develop a feed with a lower solids content to reduce the likelihood of plugging in the longer duration WSEP runs.

During processing, melt level was best detected by pot internal thermocouple readings. The view into the pot through a window in the transition piece between the calciner and the pot was impaired by the formation of a thin layer of B_2O_3 powder on the glass.

A ring of calcine and B_2O_3 powder about 3/4 in. thick formed around the top of the pot. The ring was formed at this point by the partial fusion of the flux. Had the flux

TABLE 6. *Feed Compositions for Developmental Spray Solidifier Runs*

<u>Run No.</u> <u>Waste Type</u>	DSC-35 PW-4m	DSC-36 Fast Reactor
<u>Chemicals</u>		
Fe ⁺³	0.128M ^(a)	0.372M ^(b)
Cr ⁺³	0.012	0.046
Ni ⁺²	0.051	0.183
Al ⁺³	0.001	---
Na ⁺	0.342 ^(c)	0.732 ^(d)
SiO ₃ ⁻²	0.01	---
PO ₄ ⁻³	0.003	0.06
NO ₃ ⁻	2.428	4.42
H ⁺	0.537	0.18
OH ⁻	---	0.1
<u>Fission Products</u>		
MoO ₄ ⁻²	0.161	0.30
Sr ⁺²	0.036	0.040
Ba ⁺²	0.041	0.068
Rb + Cs (used K ⁺)	0.092	0.229
(Y + RE) ⁺³	0.274	0.528
ZrO ⁺²	0.143	0.222
Ru ⁺⁴	0.0041 ^(c)	0.008 ^(f)
Rh (used Co ⁺²)	0.013	0.060
Ag ⁺	0.0016	0.019
Cd ⁺²	0.0025	0.010
Te ⁺⁴	0.014 (used SO ₄ ²⁻)	0.034
Pd (used Ni ⁺² , included above)	0.043	0.16
Tc (used MoO ₄ ⁻² , included above)	0.031	0.06

TABLE 6. (Contd)

<u>Run No.</u>	<u>DSC-35</u>	<u>DSC-36</u>
<u>Waste Type</u>	<u>PW-4m</u>	<u>Fast Reactor</u>
<u>Fission Products</u>		
Sn ⁺⁴	---	0.011
Sb ⁺³	---	0.003
SeO ₄ ⁻²	---	0.005
<u>Additives</u>		
Fe(NO ₃) ₃	---	2.17
ZnO	0.50	---
Ca(OH) ₂	0.65	---
H ₃ PO ₄	---	5.66
NaOH	---	2.80
B ₂ O ₃ (added as solid to melt pot)	2.0	---

- (a) Nominal PW-4m Fe concentration is 0.05M. Excess is Ru substitute.
- (b) Nominal fast reactor waste Fe concentration is 0.16M. Excess is Ru substitute
- (c) Nominal PW-4m Na concentration is 0.1M. Excess Na is from using Na₂MoO₄ • 2H₂O for Mo addition.
- (d) Nominal fast reactor waste Na concentration is 0.1M. Excess Na is from using Na₂MoO₄ • 2H₂O for Mo addition, Na₂SnO₃ for Sn addition, and Na₂SeO₄ for Se addition.
- (e) Nominal PW-4m ruthenium concentration is 0.082M.
- (f) Nominal fast reactor waste ruthenium concentration is 0.22M.

TABLE 7. Summary of Developmental Spray Solidifier Runs

Run No.	<u>DSC-35</u>	<u>DSC-36</u>
Feed Type	PW-4m	Fast Reactor
Feed Concentration, liters/tonne	473	912
Additives	B ₂ O ₃ (solid add to pot), Ca(OH) ₂ , ZnO	Fe, Na, PO ₄
Feed Total Acidity, <u>M</u> H ⁺	Neutral pH=7	10.5
Feed Consumed, liters	320	99 [•]
Average Feed Rate, liters/hr	17.9	14.1
Calclner Furnace Temperature, °C	700	700
Calclner Vacuum, in. of water	5	5-8
Melter Furnace Tempera- ture, °C	---	1200-1250
Atomizing Gas	air	air
Atomizing Gas Flow, ft ³ /min (STP)	6.3	6.6
Filter Blowback Pressure, psig	30	30
Filter Pressure Drop, end of run, in. of water	5	4
Melt Storage Pot Temperature, °C	950	800
Solid B ₂ O ₃ Added to Melt Pot, kg	35.5	0
Product Collect, kg	78.6	34
Product Density, g/cm ³	3.4	3.5
Particulate DF (feed to condensate)	600	Not Available
Ruthenium Volatilized, % of Total Feed Ruthenium	<1	<2

been added in a larger particle size (1/16 to 1/8 in.), it probably would not have adhered to the pot wall.

Less than 1% of the ruthenium was released from the neutralized feed during the run.

Following the run, the product pot was sectioned longitudinally. Neither the pot wall nor the thermowell was noticeably corroded. The melt filled the pot evenly to a depth of 31 in. The product varied in appearance from a black crystalline material at the bottom of the pot to a black glassy solid at the top. All the product was hard and brittle, much like glass.

Fast Reactor Waste Solidification Studies - J. D. Moore

A developmental spray solidifier Run DSC-36 was made using a simulated waste equivalent to that resulting from the reprocessing of a fast reactor fuel (100,000 MWd/tonne at 200 MW/tonne). The purpose of the run was to observe feed flow stability, melt viscosity, and ruthenium retention in the solid product.

Iron, sodium, and phosphoric acid to give an M^{+}/P of 2.5 were added to the simulated waste to give a chemical composition (see Table 6) similar to the PW-4m composition used in WSEP Runs SS-9 and SS-10. Operating conditions and results of DSC-36 are summarized in Table 7.

During the 7-hr run, feed flow to the calciner was smooth and virtually no calcine adhered to the calciner inner walls. A steady-state filter pressure drop of only 4 in. of water was maintained with a filter blowback pressure of 30 psig. The melt flowed freely from the melter

freeze valve at a melter furnace temperature of 1200 to 1250 °C. The product was a dark gray solid crystalline material with a melting point of 860 °C and a density of 3.5 g/cm³.

Less than 2% of the ruthenium was released from the feed during the run. This low volatilization rate is not characteristic of other tests with high-acid feeds (10.5M total acid for DSC-36). Usually, about 30 to 50% of the ruthenium is released. The reason for the low volatilization has not been determined at this time.

WSEP AUXILIARY STUDIES

SAMPLING OF THE PROCESS OFF-GAS J. N. Hartley

Gas sampling techniques were initiated during spray solidification Run SS-9 in an attempt to provide engineering-scale data that will permit better definition of the type, form, and extent of radioactivity present in the process off-gas. The final process off-gas leaving the caustic scrubber was sampled for both particulate and volatile forms of radioactivity by routing a side stream of the gaseous effluent through first a glass fiber filter and then a 2M NaOH solution. During Run SS-10, the process off-gas was sampled downstream of the solidifier condenser as well as downstream of the scrubber.

During phosphate glass Runs PG-7, PG-8, PG-9 and PG-10, the final process off-gas leaving the scrubber (TK-118), and gaseous effluents from the solidifier condenser (E-111), the auxiliary evaporator condenser (E-113), and the acid fractionator condenser (E-115) were sampled for both particulate and volatile forms of radioactivity by routing a

side stream of the gaseous effluent through first a glass fiber filter and then a 2M KOH scrubber. The condenser off-gas sampling system and the final off-gas sampling system are shown in Figures 2 and 3, respectively.

The condenser off-gas sampling equipment (shown again in Figure 4) consists of three separate samplers on one rack with a common vacuum header. Each sampler includes a flowmeter (0 to 1.1 ft³/min at STP) and a vacuum gauge. The sample packages which are used in conjunction with the samplers consist of a 25 mm Gelman Type A glass filter to collect particulates, a gas disperser, and a 2M KOH scrubber to absorb the volatile components such as RuO₄.

During Runs PG-9 and PG-10, gas samples were also taken of the process off-gas leaving the high efficiency filter (- 1 2) These samples were taken to determine the efficiency of the filter in decontaminating the off-gas.

The average concentrations of radioactivity in the solidifier condenser off-gas, evaporator condenser off-gas, fractionator off-gas, and the final process off-gas leaving the scrubber during Runs SS-9, SS-10, PG-7, PG-8, PG-9, and PG-10 are summarized in Table 8.

RUTHENIUM DECONTAMINATION ACROSS THE WSEP AUXILIARY EVAPORATOR AND EVAPORATOR CONDENSER DURING SPRAY SOLIDIFICATION RUNS SS-4 THROUGH SS-10 - J. N. Hartley

Based on Ru-106 concentrations in the evaporator bottoms and evaporator overheads, instantaneous ruthenium decontamination factors (DF's) were calculated for the auxiliary evaporator. Figure 5 is a plot of ruthenium DF's versus overhead acidity during Runs SS-4 through SS-10 including SS-8 and SS-10 feed

FIGURE 2. Condenser Off-Gas Sampling System

FIGURE 3. *Final Process Off-Gas Sampling System*

Neg 0692265-6

FIGURE 4 Condenser Off-Gas Sampling Equipment

TABLE 8. Radionuclides in the Process Off-Gas

Run No.	Process Condenser (E-111)			Evaporator Condenser (E-113)			Fractionator Condenser (E-115)			Final Process Offgas		
	Offgas, $\mu\text{Ci}/\text{cm}^3$			Offgas, $\mu\text{Ci}/\text{cm}^3$			Offgas, $\mu\text{Ci}/\text{cm}^3$			From Scrubber, $\mu\text{Ci}/\text{cm}^3$		
	Ru-106	Cs-137	CePr-144	Ru-106	Cs-137	CePr-144	Ru-106	Cs-137	CePr-144	Ru-106	Cs-137	CePr-144
SS-9	---	---	---	---	---	---	---	---	---	1.0×10^{-8}	---	1.3×10^{-9}
SS-10	NA	NA	NA	---	---	---	---	---	---	1.7×10^{-7}	1.1×10^{-9}	3.0×10^{-7}
PG-7a	NA	NA	NA	NA	NA	NA	NA	NA	NA	5.7×10^{-8}	6.0×10^{-9}	7.2×10^{-6}
b	3.2×10^{-3}	2.1×10^{-5}	4.5×10^{-4}	2.9×10^{-5}	9.8×10^{-7}	3.6×10^{-4}	NA	NA	NA	1.3×10^{-6}	4.4×10^{-7}	2.5×10^{-7}
PG-8a	4.8×10^{-4}	7.5×10^{-5}	2.3×10^{-5}	5.6×10^{-5}	4.5×10^{-7}	1.9×10^{-5}	2.4×10^{-7}	9.3×10^{-8}	5.0×10^{-7}	4.1×10^{-8}	6.5×10^{-8}	3.1×10^{-8}
b	2.1×10^{-2}	2.7×10^{-4}	8.8×10^{-4}	4.1×10^{-5}	4.5×10^{-7}	1.9×10^{-5}	3.0×10^{-8}	5.5×10^{-9}	4.1×10^{-7}	7.2×10^{-8}	3.3×10^{-8}	4.7×10^{-8}
PG-9 & 10	NA	NA	NA	NA	NA	NA	NA	NA	NA	2.5×10^{-8}	8.9×10^{-9}	6.7×10^{-9}

a = Feed off to Solidifier

b = Feed on to Solidifier

NA = Not Available

FIGURE 5. WSEP Auxiliary Evaporator Instantaneous Ruthenium Decontamination Factors Versus Evaporator Overhead Acidity

preparation (acid stripping). There is an apparent correlation between ruthenium DF and overhead acidity $[DF \approx 400 (\underline{M}HNO_3)^{-2.5}]$ during feed preparation with no feed on to the solidifier. However, as can be seen in Figure 5, during spray runs with feed on to the solidifier, there is no apparent correlation of DF and overhead acidity. The better ruthenium DF's that are attained when radioactive feed is not being fed to the solidifier (at the same overhead acidity) and the occurrence of a nearly linear accumulation of ruthenium in the acid fractionator with feed on to the solidifier indicate that the main contamination of equipment downstream of the solidifier is a result of the noncondensable gas stream from the solidifier. Improved decontamination of the solidifier condenser off-gas is necessary to improve decontamination by the auxiliary systems.

EVALUATION OF SOLIDIFIED WASTE PRODUCTS

PRODUCT MEASUREMENTS, TESTING, AND STORAGE

Comparison of Leach Rates - J. E. Mendel

Product measurements are being made on (1) laboratory-prepared nonradioactive products which have been subjected to varying thermal histories and (2) on samples of highly radioactive products prepared under process conditions in WSEP. A comparison of the leach rates of phosphate glass products of the two types, made from waste having the PW-2 composition, is shown in Figure 6. The leach rates for the radioactive products were somewhat lower than for the nonradioactive products. The differences can be explained in terms of thermal history effects.

FIGURE 6. Comparison of Leach Rates

The leach rates were measured at 25 °C using the procedure developed by Paige.* Phosphate glass PW-2 product has the following nominal composition (wt%):

Fe ₂ O ₃	10.3	ZrO ₂	3.3
Cr ₂ O ₃	0.5	MoO ₃	2.3
NiO	0.6	RuO ₂	1.2
Na ₂ O	8.4	Co ₃ O ₄	0.2
U ₃ O ₈	0.8	SrO	0.5
SiO ₂	0.2	BaO	0.9
K ₂ O	0.6	P ₂ O ₅	64.6
RE ₂ O ₃	5.7		

The samples of radioactive product were taken by momentarily inserting a small stainless steel cup into the molten melt stream as it was being discharged from the melter. These samples, which were essentially air quenched, formed the most nonleachable form of phosphate glass PW-2 product that has been *tested*. The results obtained with laboratory-prepared phosphate glass PW-2 products indicated that thermal treatment could grossly

affect leach rates. In the test; shown in Figure 6 the product cooled at 100 °C/hr was more leachable than that cooled at 30 °C/hr. However, in other tests in which the melt was held molten for longer times or at higher temperatures the effect was not as apparent. Storage temperatures produced the largest effects. The product devitrified when stored at 600 °C and became much more leachable. In another test, phosphate glass PW-2 product did not devitrify when held 8 weeks at 500 °C and initial leach rates

* B. E. Paige. Leachability of Glass Prepared from: Highly Radioactive Calcined Alumina Waste, IDO-14672. Phillips Petroleum Company, NRTS, Idaho Falls, Idaho., Feb. 1, 1966.

were 4×10^{-6} g/cm²/day: a value that indicates the product quality was as good as that of the product stored at 100 °C.

The leach rates of the nonradioactive samples are based on analysis of the sodium content of the leach water, and are calculated:

$$\frac{\text{fraction Na leached/day}}{\text{sample area, cm}^2/\text{sample weight, g}} = \text{g/cm}^2/\text{day}.$$

Because sodium is known to be somewhat preferentially leached, it was of interest to compare overall weight loss during leaching with that calculated from the leach rates.

Run No.	Sample Weight, g	Surface Area, cm ²	40 Weeks Leaching	
			Measured Weight Loss, g	Weight Loss Based on Cumulative Leach Rates, g
4	21.6006	30.18	0.0363	0.0751
5	17.0200	27.22	0.0198	0.0283
8	23.9066	32.72	7.883	12.286

The weight losses based on leach rates were 1.5 to 2 times greater than the actual weight losses. The devitrified phosphate glass (Run 8) had a strong skeletal structure and did not change shape or fall apart during leaching even though one-third its original weight was removed.

Solids Storage Engineering Test Facility (SSETE) - R. J. Thompson

The solidified waste containers from WSEP Runs PG-6 and SS-4 were installed in environmental test pods, and testing was initiated. Table 9 shows a status and summary of filled waste containers and environmental test pods

TABLE 9. *SSETF Environmental Test Summary*

Pot	Date Transferred to SSETF	Date Testing Started	Pod Environment	Approximate Temperature, °C			General Remarks
				Pod	Pot Wall	Pot C	
PG-4	1/69	---	---	---	--	---	PG-4 was found to have a defective end cap weld which must <i>be</i> repaired before testing can continue.
SS-4	8/69	9/69	Water	82	85	106	hunt
PG-6	9/69	9/69	Water	90	85	166	None
PC-6	9/69	--	---				In Assembly Stage.

that have entered the SSETF test program. One waste container (PG-4) was removed from immediate test because of a defective end cap weld. The waste container from Run PC-6 has been moved into SSETF and is being readied for environmental testing. PC-6 will be the first high temperature air stored container.

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