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NUCLEAR SAFETY QUARTERLY REPORT
FEBRUARY, MARCH, APRIL, 1970
FOR
USAEC DIVISION
OF REACTOR DEVELOPMENT AND TECHNOLOGY

May 1970

AEC RESEARCH & DEVELOPMENT REPORT

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By

The Staff of Battelle - Northwest

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FOREWORD

This report is the thirteenth of a series in which Pacific Northwest Laboratories reports its nuclear safety-related studies being performed for the USAEC Division of Reactor Development and Technology.

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BNWL-433	January, February, March	1967
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1. SUMMARY

ENGINEERED SAFETY SYSTEMS STUDIES

Containment Systems Experiment (CSE)

The series of five large scale demonstration runs to evaluate the performance of air cleaning systems in removing airborne fission products in a containment vessel was completed with two runs during the quarter. Run A-16 evaluated system performance during an extended duration release of fission product aerosol material. Run A-17 evaluated system performance at a higher flow rate than in preceding runs.

Results from these runs were consistent with predictions of simple mathematical models based on a well mixed gas volume for about 2 hr after the start of air cleaning system operation. After 2 hr, the removal rate of fission products was slower. The early removal rate was slightly slower than predicted for most fission products; however, the elemental iodine was removed more rapidly than predicted. The more rapid removal of elemental iodine is believed to result from the effect of higher gas phase velocity induced by the air cleaning loop operation. The higher gas velocity resulted in a more rapid deposition of elemental iodine by natural processes.

A series of small scale experiments was performed in a 905-liter stainless steel tank to evaluate the gas-liquid distribution of iodine in base-borate containment spray

solutions. Such data are needed for predictions of spray system effectiveness in removing airborne elemental iodine in containment. Contrary to theoretical predictions, the distribution coefficient at the instant of iodine release was found to increase with increasing liquid phase iodine concentration. In all cases, values were sufficiently high (~ 5000) to cause negligible liquid phase transfer resistance.

Additional blowdown experiments were completed in which void fraction data in the outlet pipe were obtained for comparison with high speed photographic records of bubble formation in the outlet.

PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

Crack Detection in Pressure Piping by Acoustic Emission

Study of acoustic emission produced by the fatigue failure process in metal has resulted in tangible evidence that the nucleation of a macrocrack is identifiable by its acoustic emission. The same work also produced evidence of the flaw size resolution attainable in carbon steel by acoustic emission - i.e., microcracking produced readily detectable emission.

Papers on acoustic emission were presented at three technical group meetings during this report period.

FIXATION OF RADIOACTIVE RESIDUES

To date, 29 radioactive demonstrations in WSEP have been completed with simulated wastes which contained approximately 45 MCi of mixed radionuclides being converted to solids.

During the report period, approximately 4.5 MCi of radioactivity were processed during the 11th and 12th spray solidification runs. Spray solidification Run SS-11 processed a simulated Purex Waste typical of that which would result from the reprocessing of a LMFBR core fuel (100,000 MWd/tonne at

200 MW/tonne) and marked the completion of WSEP spray solidifier runs using a high-temperature platinum melter. Spray Solidification Run SS-12 (IPM) was successfully completed using the Battelle-Northwest developed in-pot melting technique. During this run, spray calcined PW-4m waste was melted with a borosilicate glass frit directly in the final storage container without the need for a platinum melter. Only 1.9% of the ruthenium in the processed feed was volatilized from the spray solidifier during Run SS-12 (IPM) which verifies the results of developmental runs which indicated that the ruthenium volatilization from the spray solidifier was reduced significantly when phosphoric acid was not added to the feed.

A new method for analyzing ^{106}Ru in the spray solidifier and in the phosphate glass solidified waste products was developed successfully. The technique involves the direct gamma scan of the solid samples and (unlike previous dissolution techniques) routinely gives high ruthenium recoveries.

HEAVY SECTION STEEL TECHNOLOGY PROGRAM

Irradiation Effects on the Fracture of Heavy Section Pressure Vessel Steels

Irradiation of 1-in.-thick transverse orientation base material and submerged arc weldment specimens have been scheduled for the ETR-M3 loop during Cycles 108, 109A, 111, and 112. Specific gamma heat experiments in the ATR have been completed, which will enable the final design and construction of the 4-in.-thick specimen facility. The fatigue crack propagation rate in irradiated specimens may be double that of unirradiated material.

2. ENGINEERED SAFETY SYSTEMS STUDIESCONTAINMENT SYSTEMS EXPERIMENT - G. J. RogersFission Product Transport Studies - R. K. Hilliard,
J. D. McCormack, and L. F. ColemanLarge Air Cleaning System Experiments - J. D. McCormack

Tests were continued of air cleaning systems operating within a containment vessel under loss-of-coolant accident conditions. Two runs are reported. Run A-16 involved a 2-hr linear fission product simulant release. For Run A-17, the air flow was increased from a nominal 1000 to 1850 ft³/min. The experimental conditions are summarized in Table 2.1. This completes the planned series of five air cleaning tests.

TABLE 2.1. Summary of Run Conditions

	A-16	A-17
Date	1-20-70	3-10-70
Containment Atmosphere	Saturated Steam-Air	Saturated Steam-Air
Temperature, °F	245	246
Pressure, psia	48	48
Loop flow, actual ft ³ /min	1000	1820
Test Duration, hr	45	45
Loop Components (in order of flow)	Condenser, $\Delta T \sim 2$ °F Demister 18-in. diameter Prefilter Absolute Particu- late Filter 3 Charcoal Beds in Series, 1-in.-thick each	Condenser, $\Delta T \sim 1$ °F Demister 2 × 2 ft, square - Absolute Particu- late Filter 3 Charcoal Beds in Series, 1-in.-thick each
Aerosol Release I ₂	60 min, 104 g	10 min, 100 g
duration and Cs	120 min, 215 g	10 min, 2 g
amount delivered UO ₂	120 min, ~ 1 g	20 min, ~ 1 g
to vessel		
atmosphere CH ₃ I	10 min, 5 g	10 min, 5 g

Run A-16 tested the effectiveness of the air cleaning loop for fission product simulant removal during conditions of an extended FP release. The flow was established through the test loop just before commencement of aerosol release. The iodine was released at a uniform rate for 50 min and all iodine release was stopped at 60 min. The cesium was released at a linear rate for 120 min. The concentrations in the main gas space were plotted as a function of time in Figure 2.1 and 2.2. It appears that not all of the released aerosol was uniformly distributed throughout the gas space and that some iodine and cesium were deposited rapidly near the point of release in the drywell.

If the release rate is constant and the released mass is uniformly dispersed throughout the gas space, Equation (1) can be used to solve for the total mass released.(1)

$$W_o = \frac{\lambda V t_R C_g}{1 - e^{-\lambda t}} \quad (1)$$

where W_o = total mass released in time t_R , g
 A = overall removal rate constant, min^{-1}
 V = gas volume, m^3
 t_R = time duration of release, min
 t = time after start of release, min
 C_g = concentration in gas space at time t , g/m^3

By using Equation (1) it is calculated that 48 g of cesium and 22 g of iodine were released to the main room gas space. The uranium release was not uniform as evidenced by the decreasing concentration during the release period. The removal of material from the gas space by the loop can be found from the observed concentration half-times and by allowance for the natural removal rates, as determined from other experiments during times where the air loop was not operating. This performance is indicated in Table 2.2.

FIGURE 2.1 odine Concentration in Main Gas Space - CSE Run A16

FIGURE 2.2. Cesium and Uranium Concentration in Main Gas Space - CSE Run A16

TABLE 2.2. Aerosol Behavior in CSE Run A-16^(a)

Form	Concentration Half-Time, min			$C/C_{\max}$ at $t = 1000 \text{ min}$
	Measured, Natural, and Loop	Natural Only (Average of 8 CSE Runs)	Calculated, Loop Only	
Elemental Iodine	7.3	15	14.3	1.8×10^{-3}
Methyl Iodide	16	-	16	1.7×10^{-3}
Cesium	9.3	47	11.5	8×10^{-5}
Particulate Iodine	15	36	25.7	3×10^{-4}
Uranium	~16	31	33	
Average			20.1	

$$a. \text{ Expected } t_{1/2} = \frac{0.693}{F} V = \frac{(0.693)(21,000)}{1020} = 14.2 \text{ min}$$

The observed removal rates are in reasonable agreement with the calculated removal with the exception of particle associated iodine forms, for which the net removal was slower than expected. This may in part be induced by a slow continued release from the delivery line during the balance of the cesium release. The uranium half-time is uncertain because of the low counting rates. The 16-min half-time for methyl iodide is consistent with a bed efficiency of 88%. The charcoal beds were moisture equilibrated before use, and this is known to reduce the bed efficiency.

After the test, the loop inventory was determined and the amount of iodine and cesium on the various components is given in Table 2.3 for Runs A-16 and A-17. It is obvious that much of the iodine in Run A-16 was particulate (probably $C_s I$), as can be seen from the airborne forms in Figure 2.1 and from the large amount on the demister and filter components. This is thought to have been induced by the large amount of cesium released simultaneously with the iodine. The amount of iodine recovered from the loop after Run A-16 was higher than

TABLE 2.3. Post Run Loop Inventory, (percent of total material recovered on Loop Components) (a)

	Run A-16		Run A-17	
	Cs	I	Cs	I
HEX Condensate	11	9.2	4.8	4.0
Demister	51.6	30.6	52.2	0.7
Prefilter	2.9	1.3	-	-
HEPA	34.3	28.2	43	26.8
First Charcoal Bed				
1st half	-	16	-	40.9
2nd half	-	6.1	-	12.9
Second Charcoal Bed				
1st half	-	4.0	-	5.4
2nd half	-	2.5	-	4.1
Third Charcoal Bed				
1st half	-	1.2	-	2.6
2nd half	-	0.76	-	2.1
Total, g	31	30.5	0.95	22.7
Calculated, g	30 ^(b)	23 ^(c)	0.77 ^(d)	14.4 ^(d)

- a. Based on the total material recovered from the loop.
- b. Based on C_{max} and 120 min uniform release.
- c. Based on C_{max} and 60 min uniform release.
- d. Based on concentration at 30 min and removal rate at 30 to 60 min.

expected from the calculated release. This may have been caused by the desorption of iodine from the surfaces of the containment vessel with subsequent recovery by the loop. Desorption could be expected as a result of the decreasing iodine concentration and possibly as a result of decreasing humidity.⁽²⁾ Alternatively, the release to the containment vessel may not have been linear, and the estimation of the total quantity released to the main room may be low.

The efficiency of the various loop components was measured several times during the run by sampling from the loop with Maypacks. Table 2.4 summarizes the decontamination factors obtained by the several measurements. The table shows that the loop performance remained essentially constant during the first two hr.

Run A-17 was similar to A-15, reported previously, (3) except that the loop flow was maintained at $1820 \text{ ft}^3/\text{min}$ rather than $1000 \text{ ft}^3/\text{min}$. Allowing for steam condensed by the HEX, the flow was 5.3 main room volumes per hr. The simulants were released into the steam-air atmosphere. Samples were taken to establish the removal resulting from natural processes, and then the loop flow was started. The charcoal beds had been saturated with moisture prior to the aerosol release. Figures 2.3 and 2.4 give the main room concentrations. Table 2.5 presents the observed half-times.

Reasonable agreement with the loop removal and that calculated on the basis of a well mixed main room volume was obtained, with the observed removal being slightly slower than predicted, except for elemental iodine, which was removed more rapidly from the gas space. The methyl iodide removal rate indicated a charcoal bed efficiency of 72%. Previous CSE tests at $1000 \text{ ft}^3/\text{min}$ with moisture saturated charcoal beds have given 77 to 88% efficiencies. The increased flow through the bed did not reduce the efficiency quite as much as anticipated from the commonly acceptable expression.

$$f = \exp (-kd/v) \quad (2)$$

which relates the fraction not removed, f , bed depth, d , and gas velocity, v . Estimates of the decontamination factors for the filter loop components are given in Table 2.6 at several run times.

TABLE 2.4. Individual Loop Component Decontamination Factors, (a) Run A-16

Run Time, min	30			85			110			180		
	Cs	I	$\frac{\text{CH}_3\text{I}}{3}$	Cs	I	$\frac{\text{CH}_3\text{I}}{3}$	Cs	I	$\frac{\text{CH}_3\text{I}}{3}$	Cs	I	$\frac{\text{CH}_3\text{I}}{3}$
HEX + Demister	3	1	1	4	1	1	4	1	1	3	1	1
Prefilter	3	4	1	1	2	1	1	2	1	1	2	1
HEPA	>200 (b)	18	1	1000	3	1	700	2	1	>15 (a)	1	1
First Charcoal Bed	-	10	2.2	-	15	2	-	20	2	-	10	2
Second Charcoal Bed	-	6	5	-	8	4	-	10	3	-	>2 (b)	1
Third Charcoal Bed	-	-	-	-	-	-	-	-	-	-	-	-
Overall (c)	>2000 (b)	4300	53	4500	1000	30	2900	>1000 (b)	20	>40 (b)	>100 (b)	30

a. Efficiency = $1 - \frac{1}{DF}$; DF = concentration in/concentration out.
b. Measured downstream concentration was less than the blank.
c. Best estimate of all components.

FIGURE 2.3. Iodine Concentration in The Main Gas Space - CSE Run A17

FIGURE 2.4. Cesium and Uranium Concentration in The Main Gas Space - CSE Run A17

TABLE 2.5. Aerosol Behavior in CSE Run A-17^(a)

Form	Concentration Half Time, min		
	Natural Only	Natural Plus Loop	Loop Only, Calculated
Elemental I ₂	13	3.8	5.4
Methyl Iodide	-	11	11
Cesium	33	7.7	10
Particulate Iodine	25	6.3	8.7
Uranium	41	8.3	10.4
Average			9.2

a. Expected $t_{1/2}$ for loop = $\frac{0.693 F}{V} = 7.9 \text{ min.}$

To summarize, the aerosol behavior during both Runs A-16 and A-17 was consistent with a constant fraction removed from a well-mixed volume for about 2 hr after release, at which time the removal rates slowed. The initial removal was slightly slower than would be predicted by $\lambda = \text{loop flow/main room volume}$, except for elemental iodine which was removed more rapidly than expected in Run A-17. This may be explained by increased natural removal rates caused by the loop flow assisting the natural convection velocities within the containment vessel.

TABLE 2.6. Loop Decontamination Factors, CSE Run A-17

Time ^(b)	Decontamination Factor ^(a)								
	t = 35 min			t = 60 min			t = 80 min		
	Cs	I	CH ₃ I	Cs	I	CH ₃ I	Cs	I	CH ₃ I
HEX	1.2	1.3	-	1.2	1.6	-	1.2	1.4	-
Demister	6.2	1.3	-	3.7	1.1	-	2.4	2.5	-
HEPA	44	1.6	-	40	85			>13	
First Charcoal Bed		470	1.7			1.7	-		1.7
Second Charcoal Bed		>7							
Third Charcoal Bed									
Overall ^(c)	27,000	>8000	12	1900	>120	10	520	>180	7.5

a. Efficiency = $1 - \frac{1}{DF}$, DF = concentration in/concentration out.

b. Loop flow started at 30 min.

c. Best estimate of total DF

Iodine Gas-Liquid Partition - L. F. Coleman

Gas-aqueous distribution data for iodine are necessary to mathematically model the net benefit of engineered safeguards systems such as aqueous sprays. The distribution value is relevant in two ways, (1) the magnitude of the distribution value will limit the rate of removal of iodine from the gas phase when drop saturation can exist, i.e.

$$\frac{d C_g}{d t} = - \frac{F H_{eff}}{V} C_g \quad (3)$$

where C_g = gas phase concentration
 F = spray flow rate
 V = gas volume
 t = time
 H_{eff} = instantaneous distribution coefficient
 (neglecting slow chemical reactions)

and (2) the final distribution at longer times (hours, days, weeks) where chemical reactions have tended toward equilibrium:

$$C_g = C_L / H_{eq} \quad (4)$$

where C_L = concentration in liquid phase
 H_{eq} = distribution coefficient at equilibrium

Equations (3) and (4) represent simplified time-bounding conditions; more precise descriptions for overall iodine removal would include gas, surface and solution phase rate and equilibrium data for all iodine forms present in the containment environment.

Nine experiments were conducted to evaluate the gas-liquid distribution of iodine in aqueous-base borate systems. The experiments were performed in a 3-ft-diameter, 905-liter-volume stainless steel vessel with the aqueous phase distributed on

the vessel wall to maintain isothermal conditions and a wetted vessel wall. A more detailed discussion of apparatus and procedures was presented in a previous report.⁽³⁾

Table 2.7 lists the experimental conditions for the test program. Results from two tests, E-1 and E-2, were previously reported⁽³⁾ but are included here to give a complete set of results.

Experimental Procedure

The 905-liter vessel was brought to isothermal, isobaric conditions with the contained 100 liter, pH 9.5, 3000-ppm boron, base-borate aqueous phase. Thermally regulated steam addition to internal heat transfer coils replenished heat losses.

The aqueous phase was pumped from the bottom of the aqueous pool to the top portion of the cylindrical vessel walls and distributed by a circumferential, perforated ring.

Stable elemental iodine traced with ^{131}I was introduced into the system by either of three methods: (1) aqueous phase iodine addition to the liquid pool, (2) solid iodine volatilized to the gas phase, or (3) solid iodine in ampoules crushed within the liquid recirculating loop.

Gas phase samples were collected directly from containment by insertion and withdrawal of Maypack samplers.⁽⁴⁾ The volume of gas sampled was 10 liters (STP). These volumes were corrected to yield the actual volumes for the particular containment conditions.

Filtered air was added to the system to replenish losses incurred by sampling.

Fifty ml volume liquid samples were removed from the liquid recirculation loop at time intervals equivalent to gas sampling times. This volume was small relative to the 100 liter pool volume and did not disrupt the gas-liquid volume ratio.

TABLE 2.7. Iodine Partition Experimental Conditions(a)

<u>Experiment</u>	<u>Temp, °C</u>	<u>Pressure psia</u>	<u>Average Iodine Concentration In Aqueous Phase, mg/ℓ</u>	<u>Release Method</u>
E 1	80	24	0.011	gas phase, vaporized elemental iodine
E 2	80	24	0.0097	liquid phase, aqueous solution of iodine added
E 3	120	49	0.010	liquid phase, aqueous solution of iodine added
E 4	120	49	0.52	liquid phase, aqueous solution of iodine added
E 5	120	49	3.55	liquid phase, aqueous solution of iodine added
E 6	120	49	0.28	gas phase, vaporized elemental iodine
E 7	120	49	0.66	gas phase, vaporized elemental iodine
E 8	120	49	28	liquid phase, crystalline iodine added to aqueous system
E 11	120	49	25	liquid phase, crystalline iodine added to aqueous system

a. All experiments used pH 9.4 to pH 9.5 base borate; 3000 ppm boron as boric acid plus NaOH. Gas and liquid volumes were 806 and 100 ℓ, respectively. Wall film liquid flow rate 15 ℓ/min.

Iodine concentrations in gas and liquid samples were obtained by gamma energy analytical techniques.

Results and Discussion

Figure 2.5 illustrates how the basic analytical data were handled. Gas phase concentration versus time curves were obtained for the individual components of the gas sampler apparatus rather than total iodine to detect any variance in iodine behavior. Inorganic iodine totals excluded the granular charcoal bed component of the Maypack which is indicative of methyl iodine concentration. Methyl iodide concentration was incidental to total gas phase iodine at early times except in Experiment E-1 where difficulties were encountered during the iodine release to containment. Total inorganic iodine values were extrapolated to time zero intercept to obtain initial distribution values.

The initial distribution value obtained by this technique would relate to the predictive case for initial spray washout rates where drop fall residence times are on the order of seconds. If H_{eff} were infinite, the maximum removal rate would be gas phase mass transfer limited. For small values of H_{eff} , where drop saturation is possible, the rate of removal from the gas phase is limited by the value of H_{eff} .

An apparent first-order solution phase reaction with a 30 to 40 min half life was observed in all the 120 °C experiments. This is illustrated in Figure 2.5, where elemental iodine was injected directly into the aqueous phase. For a gas phase release, an additional gas phase mass transfer limited rate with a much shorter half life (5 to 8) minutes would also have been observed. A first-order solution phase reaction having an approximate rate constant of $3 \times 10^{-4} \text{ sec}^{-1}$ such as the early removal curve of Figure 2.5 would not contribute markedly to the total initial removal of iodine by

FIGURE 2.5. Iodine Behavior Versus Time - Experiment E3

sprays since spray drop residence times would be on the order of seconds. However, reaction rates of this magnitude would contribute to the decontamination which could be effected by sprays over an extended period of time. In the observed cases, this intermediate removal rate process lasted for about 150 min, after which a much slower removal rate process was involved, with a removal rate constant on the order of 10^{-6} sec^{-1} .

At the longer time periods in Figure 2.5, iodine distribution on the Maypack sampler components changed to the point where an appreciable fraction of the gasborne iodine mass appeared on the charcoal paper component. At early times, the bulk of the iodine was present on the silver components. Since this behavior is observed for experiments at different iodine mass levels, it does not appear to be a sampling problem (changes in iodine distribution caused by mass loading of the Maypack components) but rather associated with different chemical forms of iodine. For the conditions investigated, the concentration of these secondary iodine species would not be influential in the overall nuclear safety problem.

Another effect which is shown in Figure 2.5 is the increase in the partition coefficient at longer times with a decrease in temperature. In addition, the partition value nearly returned to its previous value when the temperature was again raised.

Table 2.8 lists some of the data pertinent to the nine experiments performed. Exact duplication of results was not possible, and the variation in the initial partition value was as great between duplicate experiments as it was between release techniques (gas or liquid phase injection of iodine). Theoretical values for the equilibrium partition values(5)

TABLE 2.8. Summary of Partition Experiment Results (a)

Experi- ment	Temp °C	Press. psia	Liquid Conc. C _L , mg/ℓ	H _{eff} = (C _L /C _G) (b)			Apparent Specific Rate Constant at Time Interval, sec ⁻¹	
				Time Zero	200 Min	1000 Min	20-100 Min	200-1200 Min
E 1	80	24	9.9x10 ⁻³	6,400	9,400	--	2x10 ⁻⁵	--
E 2	80	24	9.7x10 ⁻³	6,800	10,000	--	3x10 ⁻⁵	--
E 3	120	49	1.0x10 ⁻²	22,000	250,000	570,000	3x10 ⁻⁴	2x10 ⁻⁵
E 4	120	49	5 2x10 ⁻¹	21,000	400,000	520,000	4x10 ⁻⁴	< 10 ⁻⁵
E 5	120	49	5 5x10 ⁻¹	5,500	83,000	--	3x10 ⁻⁴	--
E 6	120	49	2.8x10 ⁻¹	5,600	62,000	--	4x10 ⁻⁴	--
E 7	120	49	6.6x10 ⁻¹	42,000	500,000	--	4x10 ⁻⁴	< 10 ⁻⁵
E 8	120	49	28	56,000	1,280,000	2,800,000	3x10 ⁻⁴	< 10 ⁻⁵
E 11	120	49	25	140,000	400,000	--	3x10 ⁻⁴	--

a. Conditions: 100 l liquid phase, 806 l gas phase, pH 9.4 to 9.5, 3000 ppm boron, recirculated liquid on vessel walls at 15 l/min flow rate.

b. G = total iodine in aqueous phase, C_G = total inorganic iodine in gas phase.

would be about 3×10^9 , 6×10^6 and 1×10^5 at the three iodine concentration ranges investigated.

The value of the zero-time partition coefficient, H_{eff} , is plotted as a function of iodine concentration in the liquid phase in Figure 2.6. Contrary to theory,⁽⁵⁾ the coefficient increased with increasing liquid concentration. In all cases, however, the values were sufficiently high (>5000) to cause liquid phase transfer resistance in falling drops to be negligible and to allow rapid removal of inorganic iodine from containment gas spaces by base-borate sprays. Continued removal, though at a slower rate, would occur because of the chemical reactions within the liquid. After several hours of recirculation, the concentration of inorganic iodine in the gas space would be reduced by about a factor of 10 from the value obtained by the initial fresh spray period.

Coolant Blowdown Studies - R. T. Allemann, W. C. Townsend, A. S. Neuls, M. E. Witherspoon and A. J. McElfresh

A reactor simulator vessel with a volume of 150 ft³, designed for operation at 600 °F and 2750 psig is being used in experiments to study loss-of-coolant accidents. The objectives of the experiments include testing the validity of mathematical models used for predicting the consequences of a loss-of-coolant reactor accident. The vessel is mounted in a steel framework with the discharge nozzle directed horizontally. The blowdown apparatus is instrumented to measure internal pressures, temperatures, liquid level, void fraction of the fluid in the exit duct, mass of fluid remaining in the vessel, thrust reaction forces, shell stresses, and forces on reactor dummy core structures (Figure 2.7).

During this reporting period the mid-nozzle (Nozzle B) series of blowdowns was completed and installation was begun

FIGURE 2.6. Iodine Partition Value Versus Concentration

FIGURE 2.7. CSE Blowdown Test Vessel

of a dummy core which is designed to model the internal geometry of a pressurized water reactor.

Void Fraction-Quality Measurements

The void fraction in the outlet pipe has been observed to increase with nozzle size and the associated increase in quality is the probable cause of the decrease in mass velocity with orifice size. This decrease in mass velocity appears to correlate with the cavitation number.

The reduction in the mass velocity with nozzle size⁽⁶⁾ has required a coefficient⁽⁷⁾ to correct theoretical flows to actual flows. The increase in quality is not sufficiently explained by considerations of bubble rise and separation within the vessel. Typical codes which consider bubble rise phenomena [FLASH,⁽⁸⁾ BURP,⁽⁹⁾ RELAPSE⁽¹⁰⁾] still require a coefficient to give a reasonable prediction for this special case.

The test series just completed included two runs in which the void fraction in the test section (Figures 2.7 and 2.8) was measured with a neutron densitometer. In this device, neutron moderation is used as a measure of the amount of liquid in the pipe. The neutron detector data were converted to quality values by the code DENPRØ developed for the purpose. Example of these quality data are shown in (Figures 2.9 and 2.10). The data are scattered because the digitized recording technique transmits occasional values that represent physically unrealistic situations which introduce instability into the detector output-to-quality conversion equations. However, the overall values and trends are obvious. The low quality portion of the run early in the blowdown is followed by a rapid transition to high quality flow. The low quality region is characteristic of the saturated flow regime of blowdown. The value of the quality during the saturated flow regime is about 1.5×10^{-2} for a

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FIGURE 2.9. Quality in Test Section

FIGURE 2.10. Quality in Test Section

6 in. (nominal) break and 2×10^{-3} for a 2-in. (nominal) diameter break. Although the blowdowns were started at 530 and 487 °F respectively, the predicted (on the basis of isentropic expansion) values of quality in the pipe contents are 1.7×10^{-2} and 1.4×10^{-2} respectively. Hence, the small break had a much lower quality flow than expected, and it was much lower than for the larger orifice blowdown test. Earlier data taken with the neutron densitometer had also shown that the mass velocity decrease was associated with orifice size decrease.⁽⁶⁾ These data also showed a mass velocity decrease with quality,⁽¹¹⁾ as predicted by most two-phase critical flow theories.

The experiments of Lienhard and Stephenson⁽¹²⁾ show the same effect of size as do these experiments; that is, the cavitation number is proportional to area to the -0.33 power. And since the cavitation number is

$$\sigma = \frac{2\rho(P - P_v)}{G^2},$$

it can be concluded that G_{crit} should be proportional to (area of orifice)^{-0.166}

This result is close to what has been found by direct experiment in previous work at CSE and other sites.⁽⁶⁾

The implication is that the larger break allows higher flow rates which gives more turbulence and a lower cavitation number; hence more bubble nuclei are triggered to form bubbles. The larger number of bubbles gives a higher quality which limits the mass velocity to a lower value than with smaller breaks.

Photographs taken of the fluid in the test section do not contradict this theory. High speed motion pictures taken through the window of the test section (Figure 2.8) using cross illumination showed individual bubbles moving at measurable rates (Figure 2.11) for small (0.0155 ft²) breaks.

FIGURE 2.11. High Speed Film Strips

In larger breaks no individual bubbles were seen in spite of a higher quality registered by the neutron densitometer. However, early in the blowdown for a large orifice (0.253 ft^2), a milky cloud was seen moving rapidly with the fluid. Hence, it appears that much smaller but more numerous bubbles are formed as the break size is increased. For small breaks fewer bubbles are formed which are able to grow to visible size in the time available.

To apply this conclusion to reactor pipe failures would assume that for the large breaks sufficient turbulence and nucleation sites exist for the flow rate to be limited by steam quality. Small breaks too, may act as even smaller exits, giving lower mass loss rates than static tests, because initial turbulence and nucleation sites present in a flowing loop give a higher quality at the break than in the stagnant CSE tests. However, since the quality of the flow is highly dependent upon the temperature-pressure at the outlet relative to that in other parts of the system, the determination of the fluid condition should be performed carefully. For example, emergency core cooling water, if mixed with a blowdown flow, could increase the flow rate through the break by an amount greater than the coolant addition rate. Hence, the fluid loss would be faster than without the emergency coolant addition.

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3. PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

CRACK DETECTION IN PRESSURE PIPING BY ACOUSTIC EMISSION -

P. H. Hutton and J. B. Vetrano

Introduction

The term "acoustic emission" describes a physical phenomenon which can be utilized to detect flaw formation and growth in solid materials on a real time basis. Acoustic emission consists of elastic waves produced in a solid by energy released during deformation and fracture. These waves are detectable at the material surface some distance from the source, thereby providing the means to detect the presence of flaws remotely at the time they are formed or grow.

The USAEC's Division of Reactor Development and Technology has sponsored work at Battelle-Northwest facilities since February, 1966 directed toward application of acoustic emission to detect crack formation and growth in nuclear reactor pressure boundaries. A proven system to perform this function would be a significant adjunct to nuclear safety.

Related work on the in-plant application phase has also been done at the Idaho Test Site, Idaho Falls under DRDT sponsorship.

There is a distinct potential for application of acoustic emission to the integrity surveillance portion of the LMFBR program.

Reduction in program scope and limited funding has curtailed program activity during the past quarter.

Discussion

Task 1 - Signal Analysis and Characterization

One of the major needs in acoustic emission technology at this time is substantiated criteria for interpretation of

acoustic emission information. Investigation of acoustic emission from metal fatigue has contributed a criterion point and has also provided evidence toward defining minimum detectable flaw size.

Fatigue tests of a variety of metals (2024-T3 aluminum, Type 304 SS, high-nickel steel, and mild carbon steel) in both high and low cycle fatigue has shown a consistent acoustic emission precursor to the appearance of a visible macrocrack. Although it was suspected that this indication was generated by the initial nucleation of a macrocrack, confirming information was needed. A simple notched, cantilever beam fatigue specimen of Type A-212-B carbon steel was chosen for investigation of the mechanism producing the acoustic emission benchmark. This was tested under high cycle, tension-tension fatigue. Figure 3.1 shows the specimen shape and load cycle. Also shown in Figure 3.1 is the acoustic emission response curve. The increasing emission response from 390,000 to 630,000 cycles is attributed to microcracking. The test was shut down over night at 570,000 cycles and when the test was resumed, the emission started at essentially the same level it had attained when the test was stopped. At 630,000 cycles the test was again stopped and the specimen was removed for examination. The surface was polished and lightly etched to inspect microscopically for any evidence of a macrocrack. Since none was found, it was again installed in the test fixture and cycling resumed. The emission level, however, started much lower than the previous point. This is attributed to the polishing which removed many microcrack sites from the high stress area. The abrupt increase in acoustic emission which preceded the appearance of a visible crack occurred at 705,000 cycles. The specimen was cycled an additional 11,000 cycles and then removed for metallographic examination.

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FIGURE 3.1. Acoustic Emission from Carbon Steel Fatigue Specimen

Metallographic inspection revealed an abundance of micro-cracks on both sides of the specimen near the root of the notch. Figure 3.2a shows an example of these at a depth of ~ 0.010 in. from the surface. Further investigation revealed one macro-crack at a depth of 0.125 in. into the specimen. This is shown in Figures 3.2b, 3.2c, and 3.2d. It was ~ 0.002 -in. deep and estimated to be 0.030 in. long. No other macrocracks were discovered. These data provide substantiation for the conclusion that the abrupt increase in acoustic emission observed in fatigue testing is a result of macrocrack nucleation. The data also indicate that microcracking, at least in carbon steel, is readily detectable by acoustic emission. This contributes to definition of minimum detectable flaw size.

Task 2 - Sensor Development

Task 3 - Signal Conditioning System

Task 4 - Data Analyzer and Readout System

Task 5 - Application Engineering

No significant developments have been made on any of these tasks.

Task 6 - Cooperative Work with Idaho Nuclear

Liaison has been continued with Idaho Nuclear Inc. by telephone and by a meeting at Richland on April 27, wherein program status was reviewed.

General. Papers on the subject of acoustic emission work performed under this program were presented to three technical group meetings this past quarter. These were:

- Acoustic Emission - What It Is and Its Application to Evaluate Structural Soundness of Solids, BNWL-SA-2983, P. H. Hutton, February, 1970 presented at the Central Ohio Section, ASNT, February 17, 1970 at Columbus, Ohio, and the Chicago Section, ASNT, February 19, 1970 at Chicago, Illinois.

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FIGURE 3.2a. Carbon Steel Fatigue Specimen - Emission Sources

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FIGURE 3.2b

Carbon Steel Fatigue Specimen - Emission Sources

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FIGURE 3.2c

Carbon Steel Fati

Specimen

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FIGURE 3.2d

Carbon Steel Fatigue Specimen - Emission Sources

- Acoustic Emission Applied to Determination of Structural Integrity, BNWL-SA-3147, P. H. Hutton, March, 1970
presented at the 11th Open Meeting of the Mechanical Failures Prevention Group, April 8, 1970 at Williamsburg, Virginia.

WORK PLANNED FOR NEXT QUARTER

Effort intended for the next quarter includes:

- Study of wave propagation in pipes.
- Complete study of signal attenuation versus frequency in range of significance to acoustic emission detection.
- Document feasibility assessment of using wedge-mounted lithium niobate shear sensors for high-temperature acoustic emission detection.
- Apply acoustic emission monitoring to fatigue test of 24 in. diameter pipe tee at facilities of Combustion Engineering, Chattanooga, Tenn. to confirm that macrocrack nucleation can be detected in a large specimen as well as in laboratory specimens.

4. ENVIRONMENTAL STUDIES

CALCULATION OF THE POTENTIAL ENVIRONMENTAL RADIOLOGICAL CONSEQUENCES OF REACTOR ACCIDENTS - M. M. Hendrickson

Early in 1967 the USAEC Division of Reactor Development and Technology (DRDT) asked several companies and laboratories to participate in a survey to compare methods of calculating potential environmental radiological consequences of reactor accidents. The survey consisted of a test problem for which the participants were asked to submit calculations of thyroid doses and whole body gamma doses from an airborne plume at specified distances from a hypothetical reactor accident.

Large differences were observed among the answers for the whole body gamma dose. As a result RDT authorized Battelle-Northwest to undertake a program to develop improved methods for estimating reactor accident consequences with the immediate objective of developing an improved method for the whole body gamma dose calculation.

Several of the participants used sophisticated computer codes to perform the gamma dose calculation. Some of the codes perform integrations over the dimensions of the plume and others assume the cloud to be semi-infinite in size and of uniform concentration. A detailed study of the methods used has revealed significant differences in all of the calculations. As a consequence of this study a general computer code has been developed based on insight gained from the work of the participants.

Although the main purpose of the code is the calculation of the whole body gamma dose, calculations are also included which permit accountability for the effects on the source term of the events of a hypothetical reactor accident.

Hence the computer program is divided into three main subroutines:

- Subroutine SOURCE - 1, determination of the important nuclides available for release
- Subroutine BAFFLE - 2, calculation of the nuclide release rate to the atmosphere
- Subroutine EXDOSE - 3, calculation of the external whole body gamma dose.

A description of each main portion of the code is given below. A fourth subroutine to calculate the internal dose will be added in the future.

The code is applicable to BWR's, PWR's, LMFBR's, experimental reactors, production reactors and some other nuclear facilities.

SOURCE - Initial Nuclide Inventory

The accurate calculation of radionuclide inventories is complex and depends upon many variables. A very sophisticated calculation would use a multigroup cross section model and detailed description of core components, fuel and geometry. Although such a calculation is not included in the present code, a provision is available whereby inventories obtained from such calculations may be specified (in curies) for specific isotopes. With this provision the results of any type of inventory calculation may be used with the code. Radionuclides not produced through normal fission and decay reactions (i.e. activation products) may also be supplied as input.

The inventory of radiologically significant radionuclides of most reactors can be described adequately by a simple one-group cross section calculation. An adaptation of such a computer code (RIBD - Radio Isotope Buildup and Decay) has been included in SOURCE. A multiple chain grid processor is used which calculates nuclide concentrations resulting from two fission sources (for example, ^{235}U and ^{239}Pu) with normal

down-chain decay by beta emission and isomeric transfers and interchain coupling resulting from n-gamma reactions.

The maximum number of members allowed in a chain is nine, and the maximum number of chains is 96. The program contains on tape two libraries of data for 450 nuclides each; one library is for fast reactor calculations and the other is for thermal reactor calculations. Data contained in the libraries include physical half-lives, direct yields from ^{235}U fission, direct yields from ^{239}Pu fission, neutron capture cross sections, average γ decay energies per disintegration, average β decay energies per disintegration, and branching ratios to account for various modes of transition to and from isomeric states.

When the inventory calculation is used, the code supplies the fission product inventory at the time of reactor shutdown and also for nine decay times after shutdown (specified as input). The inventory for shutdown or any one of the nine decay times may be taken as the initial inventory for the calculations to follow. The desired initial inventory (specified or calculated) is stored for use in the containment model calculation (subroutine **BAFFLE**).

The input parameters for the inventory calculation include power level, operating time, flux parameter, ^{235}U capture cross section, the initial ratio of second to first species fissions, and nine decay times.

BAFFLE - Containment Release Calculation

The rate at which nuclides are released to the atmosphere is calculated by a generalized model for describing aerosol behavior and subsequent release using fractional settling rates and fractional leak rates. The model may be used to describe release from fuel through a three-barrier (or fewer) system with filtration and settling accounted for in each

barrier. A diagram of the containment model is given in Figure 4.1. To reduce computing time the following simplifying assumptions were made:

- Instantaneous mixing in each vessel
- Fallout and leak rates may be considered constant for a short time interval

Isotopes becoming airborne from settled material come only from the mass present at the beginning of the interval

- Material released from the filters comes only from material present on the filters at the beginning of the interval.

These assumptions make the system of differential equations for nuclide concentrations in each compartment analytically solvable and of the form $\text{Mass}(t) = \sum_i A_i e^{-B_i t}$ where the summation is over all previous nuclides in the mass chain and all previous compartments. The A_i and B_i are functions of fractional leak rates, removal rates, filter efficiencies, nuclide half-lives and fractional decay yields. This portion of the code then produces a series of nuclide release rates and time points to be used in the subroutine EXDOSE.

The generalized model allows use of theoretical or experimental data for fractional leakage and settling rates by simply picking time-rate points from a graph. Thus as progress is made in the field of aerosol behavior it will be convenient to use the newly acquired data without code modification. As an example Figures 4.2 and 4.3 show how data might look that would be used as input to the code for a hypothetical accident. Data points which might be selected for use as input are marked with circles. Note that the points are not uniformly spaced but are bunched in regions of maximum change. The time rate points calculated internally by linear interpolation are printed out so they can be checked against the original data.

L_{ij} = LEAK RATE FROM COMPARTMENT (i) TO COMPARTMENT (j) , sec^{-1}
 S_i = SETTLING AND PLATEOUT RATE FOR BARRIER (i) , sec^{-1}
 F = FILTER FACTOR

FIGURE 4.1. Multiple Containment Model

FIGURE 4.2. Typical Leak Rate Data

FIGURE 4.3. Typical Settling Rate Data

If this proposed method of describing containment system action becomes generally used it will be desirable for those conducting tests to report their data in terms usable in this system, i.e. make available the results of aerosol behavior tests in terms of settling rate as a function of time for various nuclides.

The input parameters for the containment calculation include the time interval for which nuclide release rates are to be calculated, the number of time intervals to be considered, the number of time-rate points to be entered, the times for the time-rate data, the rates for the time-rate data, and filter factors. Filter factors may be specified for individual isotopes or for groups of isotopes, i.e. noble gases, halogens, volatile solids, and all remaining fission products. Fractional leak rates and removal rates describe the containment system being modeled. Unspecified rates are set to zero.

EXDOSE - Whole Body Gamma Dose Calculation

The subroutine EXDOSE calculates the external whole body gamma dose based on the release rate data generated in the subroutine BAFFLE. Dose rates may be calculated with the semi-infinite cloud assumption or for a finite cloud. The semi-infinite cloud calculation is well documented and will not be discussed here. An exact solution to the finite cloud problem has not been found. A precise and generalized numerical solution of the finite cloud dose problem is possible but is impractical because it would take unreasonably long running time on the computer and because all of the necessary data are not now available to describe exactly all of the phenomena involved. A truly precise solution would for example require the description of the activity throughout the space domain as a function of time. Neither usable models nor tests have been devised (much less conducted) which provide such detailed information.

To obtain a practical solution to the problem, the following assumptions were made:

- The receptor is on or beneath the cloud centerline at ground level downwind of the release point.
- Diffusion along the direction of travel can be ignored.
- Lateral and vertical crosswind diffusions are described by normal distribution functions.
- Cloud depletion by fallout, washout, and rainout is not included.
- The contribution to the dose rate from airborne nuclides at distances beyond three standard deviations in the vertical and lateral directions is insignificant. (For elevated release, vertical limits are $-3\sigma_z$ to $H + 3\sigma_z$ where H is height of release.)
- The contribution is insignificant beyond ± 420 meters away from the receptor in the direction of travel. (The integration limits (5) and (6) are subject to change.)

For each dose calculation the following parameters are specified: distance from release point to receptor, length of release period, length of exposure period, height of release, average wind speed in direction of travel, and the dispersion parameters σ_y and a . Three methods are available to account for atmospheric dispersion of the plume.

- Hanford model (bivariate normal mode dispersion equation with σ_y being time dependent and σ_z a function of time and atmospheric stability).
- Sutton model.

Direct input of σ_y and σ_z for each point on the integration grid in the direction of travel. (Bivariate normal mode dispersion equation is used - with σ_y and σ_z input from Pasquill or other source).

The direct input of dispersion parameters σ_y and σ_z allows an unlimited variety of atmospheric conditions to be studied.

In Models 1 and 3 the dispersion (time integral of air concentration) is accounted for by the commonly used equation

$$X = \frac{Q'}{\pi \sigma_y \sigma_z \bar{u}} \exp \left[-\frac{y^2}{2\sigma_y^2} + \frac{h^2}{2\sigma_z^2} \right]$$

(continuous point source, receptor at ground level)

The Hanford model for long-period releases is described in the 1968 edition of Meteorology and Atomic Energy pp. 380-392.

The dose integration kernel is

$$D_k(X, Y, Z, t, E) = \left(\frac{\tilde{X}}{\bar{Q}} \right) \frac{B(E, R) e^{-a(E) R}}{4\pi R^2} K(E)$$

where

D_k = dose kernel as a function of position, time and nuclide γ -energy,

$$\frac{\text{rad}}{\text{cm}^3 \text{sec}}$$

$\left(\frac{\tilde{X}}{\bar{Q}} \right)$ = instantaneous cloud concentration as a function of position

$$\left(\frac{\text{curie}}{\text{cm}^3} \right) / \text{curie released}$$

R = distance from exposure point to kernel in cloud, (cm).

B = Taylor buildup factor, a function of nuclide γ -energy (dimensionless)

a = attenuation coefficient, a function of γ -energy, (cm^{-1})

K = dose rate conversion factor, a function of y-energy

$$\frac{\text{rads}}{\text{sec}} \left(\frac{\text{cm}^2 \text{ sec}}{\text{photon}} \right) \left(\frac{\text{photon/sec}}{\text{curie}} \right) = \frac{\text{rads} \cdot \text{cm}^2}{\text{curie} \cdot \text{sec}}$$

The dose kernel is integrated over time and space to obtain the dose in rads.

For each set of data the code first calculates the dose rate per curie per cm of cloud (in direction of travel) for each of 16 energy groups. These dose rate factors involve integration over the y-z plane at each of the points on the integration grid in the direction of cloud travel. The points in the direction of travel are chosen around the exposure point to ensure accuracy in the numerical integration. The dose per cm at each point is calculated as

$$\text{Dose per cm} = \int_0^T \sum_{\text{isotopes}} \sum_{\text{energies}} \left(\frac{\text{Dose Rate}}{\text{Curie cm}} \right) (\text{Curies}) +_{ei} dt$$

where F_{ei} is the fraction of gamma energy from nuclide i in energy level e.

The dose is given by

$$\text{Dose} = \int_{x_1}^{x_2} (\text{dose per cm}) dx$$

The amount of each nuclide ("curies" in above equations) at each integration point is calculated as a function of time. The calculation accounts for chain decay with branching in a similar fashion to the initial inventory calculation of subroutine SOURCE.

The gamma energy spectrum has been divided into 16 energy groups as listed in Table 4.1. The main reason for considering 16 energy groups is that a 16 group data library is available.

TABLE 4.1. Energy Groups

<u>Group No.</u>	<u>Upper Limit Energy, MeV</u>	<u>Average Energy, MeV</u>
1	0.2	0.150
2	0.3	0.250
3	0.4	0.350
4	0.55	0.475
5	0.75	0.650
6	0.9	0.825
7	1.1	1.000
8	1.35	1.225
9	1.6	1.475
10	1.8	1.7
11	2.0	1.9
12	2.2	2.1
13	2.4	2.3
14	2.6	2.5
15	2.8	2.7
16		3.0

Much time and money has been invested in the development of this library. A single group calculation was not used since such a calculation could introduce significant error for individual nuclides.

Computer Requirements

The code has been programmed in FORTRAN V for the Univac 1108 computer operated by Computer Science Corporation at Richland, Washington. The Univac 1108 has available a magnetic core memory of 36 bit words that totals 131,072, 45 million words of magnetic drum memory, and 8 compatible UNISERVO VIIIC tape drives. The code presently uses only two tape drives

(for data storage). The size of the main subroutines (SOURCE, BAFFLE and EXDOSE) dictates that only one be in the core at a time. Therefore, the tape drives are necessary to store data from one subroutine while the next subroutine is being loaded (drum memory may also be used instead of tapes). Figure 4.4 illustrates the relation of tape drives and drums for the four main subroutines. This relation allows the subroutine to be entered in any order. For example a typical run may have the following calling sequence:

SOURCE (for inventory) - 5 to 30 sec computer time
 BAFFLE (for release) - 5 to 60 sec
 EXDOSE (y - dose for 1st meteorological condition) - 10 to 30
 EXDOSE (y - dose for 2nd meteorological condition) - 10 to 30
 BAFFLE (different release of same inventory)
 EXDOSE (y - dose for 1st meteorological condition)
 EXDOSE (y - dose for 2nd meteorological condition)
 SOURCE (new inventory), etc.

REGIONAL MODELING OF SURFACE WATER TEMPERATURE FROM PROJECTED
 POWER GROWTH - R. T. Jaske and D. E. Peterson

A report, Potential Thermal Effects of An Expanding Power Industry: Upper Mississippi River Basin, was completed in draft form during the report period. The results are summarized in Figures 4.5, 4.6, and 4.7, average and low flow cooling capacities of the Upper Mississippi River from Royalton, Minnesota to Alton, Illinois. Direct cooling capacities were simulated by means of the MAXPWR computer code. Constraints were a temperature envelope of 5 °F above natural background and incremental advected heat increases of 800 thermal megawatts (MW_t). Because of a combination of relatively low streamflow and low energy exchange with the atmosphere, November was found to be the critical month for both average and low-flow conditions, with total UMRB capacities of 61,000 MW_t and 46,000 MW_t respectively.

FIGURE 4.4. Code Flow Diagram

FIGURE 4.5. Average Direct Cooling Capacity of Upper Mississippi River

FIGURE 4.6. Low Flow Direct Cooling Capacity of Mississippi River

FIGURE 4.7. Upper Mississippi River Basin
Local Direct Cooling Capacities

In general, cooling capacities were adequate along the main stem Mississippi to assimilate and dissipate projected 1980 thermal loads. Upstream of the confluence of the Mississippi and Chippewa Rivers, in the St. Paul-Minneapolis area, low flow heat sink capacities were found to be marginal with respect to present loads in the downtown area and with respect to 1980 loads in adjacent reaches of the Mississippi, Minnesota and St. Croix Rivers. Tributary streams, including the Minnesota, St. Croix, Chippewa, Cedar-Iowa, Des Moines and Illinois Rivers have insufficient cooling capacities without ancillary cooling facilities under low flow conditions for projected 1980 and/or 1990 loads. Capacities of the Wisconsin and Rock Rivers are adequate for 1980 conditions primarily because of an expected low level increase in local generating capacity. Rock River thermal capacities are inadequate for projected 1990 loads. The lower Wisconsin River, which has a local heat sink capacity of 900 MW_t or greater 99% of the time, may remain without significant sources of thermal effluents through 1990 because the nearby central section of the Upper Mississippi could accommodate projected loads without auxiliary cooling facilities. By 1990 projected generating capacity in the Rock Island-Moline-East Moline-Davenport (Quad Cities) area would be marginal with respect to calculated low-flow cooling capacities. A similar condition holds in the Alton-St. Louis area. At the 99% probability level local capacities are 4600 MW_t in the Quad Cities, 6000 MW_t at Alton and 8100 MW_t at St. Louis. If sources of thermal effluents are not concentrated in short reaches, atmospheric dissipation would increase area capacity significantly.

Simulations of 1964 temperatures of the Missouri River from Sioux City, Iowa to St. Louis and of the Upper Mississippi River from Alton to Cairo, Illinois were completed satisfactorily during the report period. Determinations of the 5°F heat sink

capacities of the Missouri River and the lowermost 200 miles of the Upper Mississippi River are scheduled for May and June. MAXPWR simulations of thermal capacities of the Columbia-Snake River system within a temperature envelope of 2 °F are also scheduled for May and June.

Reference

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5. FIXATION OF RADIOACTIVE RESIDUES

OVERALL STATUS OF WSEP RADIOACTIVE DEMONSTRATIONS

K. J. Schneider

Table 5.1 shows the overall status of the radioactive demonstrations in WSEP as of April 30, 1970. A total of 29 demonstrations have been completed: 6 in the pot calcination process, 11 in the phosphate glass solidification process, and 12 with the spray solidification process. Approximately 45 MCi of mixed radionuclides representing waste from about 263,000 MWd of electrical power have been converted to solids. This amount of waste represents 65 days of all the nuclear power capacity in the United States as of February 1, 1970. The total on-line processing time for solidification of these wastes was also 65 days.

TABLE 5.1. Overall Status of WSEP Radioactive Demonstrations

	<u>Solidification Methods</u>			<u>Total</u>
	<u>Pot</u>	<u>Spray</u>	<u>Phosphate Glass</u>	
Runs Completed	6	12	11	29
Megacuries Solidified	4.0	21.5	19	45
Equivalent Tonnes(a) Processed	11.3	15.7	12.4	39
MWd _e Represented by Waste(b)	75,000	105,000	83,000	263,000
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	12.0	167 ^(e)
Maximum W/L in Pot	85	205	315 ^{(c)(d)}	--
Maximum Centerline Temperature in Pot, °C	940	930	780	--
Liters Solid/Tonne ^(a)	40 to 50	30 to 100	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. This value is for 6-in. diameter pots, whereas the comparable values for the pot and spray solidifiers are for 8-in. diameter pots.

d. For 8-in. diameter pots, the maximum is 195 W/l.

e. Totals to date from all processes.

SPRAY SOLIDIFICATIONLABORATORY STUDIESIn-Pot Melting Development - J. E. Mendel and J. N. Strode

The laboratory program to develop chemical flowsheets for the final two WSEP spray solidifier runs wherein the spray calcined waste will be melted directly in the final storage container (in-pot melting) is nearly complete. The effects of such parameters as flux composition, ratio of flux to waste calcine, chemical additions to the waste before calcination, and processing temperature have been examined.

For the in-pot melting runs the continuous platinum melter of the WSEP spray solidifier has been replaced with a special spool-piece containing a port for the addition of a solid fluxing agent. The solid fluxing agent is added to combine with the calcine in the correct ratio to form a monolithic product upon fusion in the stainless steel receiver, which is also the final storage vessel. The fluxing agent must have certain properties: .

- a Relatively low melting point so that fusion can be carried out in stainless steel containers.
- Low corrosivity to stainless steel.
- Ability to combine with calcine to form a relatively homogeneous solid free of void spaces.
- a Low leachability.

Compositions of the solid fluxing agents which were considered in the laboratory test program are given in Table 5.2. These were . all glass frits which were obtained commercially or, in the case of the phosphate-containing frits, prepared in the laboratory. Limited tests were also made using the compounds B_2O_3 and $NaPO_3$ for fluxing.

TABLE 5.2. Flux Compositions Used for In-Pot Melting Tests

Approximate Composition, wt%	Frit 111 (a)	Frit 111-15 (a)	Frit 1806 (a)	Frit 78-1 (a)	Frit 1415 (a)	Coke Bottle	Borosilico Phosphate Glass Frit				Phosphate Glass Frit	
							I	II	III	IV	I	II
B ₂ O ₃	26	36	30	23	18		28	30	24	26		
SiO ₂	37	32	36	20	55	73	32	34	28	30		
Na ₂ O-K ₂ O	11	10	16	1	19	16	14.5	16	26	27	8	4
ZnO	18	15									9	9
CaO	2	1.75	6		1	5						
Al ₂ O ₃			4			2	12	13	10	11	10	12
MgO	2	1.75				4						
SrO	2	1.75										
BaO	2	1.75	6									
F			2									
PbO				50								
Fe ₂ O ₃				5								
Cr ₂ O ₃				1								
TiO ₂					2							
Co ₃ O ₄					2							
NiO					2							
P ₂ O ₅							13.5	7	12	6	73	75
	100	100	100	100	100	100	100	100	100	100	100	100
Accelerated Leach Rate, % Weight Loss												
First 24 hr	0.09		4.3	11.3	5.6	0.31					3.9	
Second 24 hr	0.05		4.3	10.6	5.0	0.27					4.0	
Third 24 hr	0.02		4.1	3.6	5.5	0.24					4.1	
Overall, 72 hr	0.16		12.2	23.6	15.3	0.82					11.5	

a. Manufactured by American Porcelain Enamel Company, Muskegon, Michigan.

The laboratory tests made were of two types: batch melts of 20 to 50 g and continuous runs in 2-in. diameter pots to make about 1500 g of melt. For the batch melts, solid flux and calcine were premixed; for the continuous runs flux and calcine were added semicontinuously in two streams at the desired ratio. The calcine powder which was used was prepared from the PW-4m waste composition and was either batch calcined and ground in the laboratory or prepared in the cold developmental spray calciner. In either case the calcine particles were quite fine with more than 95% passing a 200 mesh screen. Various sizes of frit particles have been used, but in most cases the frit particles have been considerably larger than the calcine particles.

The flux chosen for in-pot melting Runs SS-12 and 13 was frit 111-15 (composition given in Table 5.2), a low-melting borosilicate glass. The decision was based principally upon considerations of potential pot corrosion and melt quality, i.e., leachability and homogeneity.

Void-free melts were obtained with B_2O_3 and $NaPO_3$ fluxing, but the products were undesirably leachable and, in the case of $NaPO_3$, the melt was unacceptably corrosive to stainless steel. Phosphate glass frits I and II yielded homogeneous melts at 950 °C containing 30 wt% PW-4m in laboratory batch tests. The melt proved too corrosive for further consideration however. In a continuous run in a 2-in. pot made of 304L stainless steel the pot wall corroded through after 24 hr at 950 °C. Of the four borosilic-phosphate glass frits which were examined briefly, composition I yielded the most homogeneous melts, but temperatures above 950 °C were required (the arbitrary upper operating temperature for in-pot melting in 304L stainless steel pots). Coke bottle glass, a soda lime silicate formulation, also required temperatures above 950 °C to obtain void

free melts. Melts with coke bottle glass also had 2 to 3 vol% of a second phase which floated on the glass melt and solidified to a soluble yellow microcrystalline phase.

The borosilicate frits were studied in most detail. Melts of calcine plus the lead-containing frit (frit 78-1) were about an order of magnitude more corrosive to stainless steel than melts made with borosilicate frits not containing lead. Since frit 78-1 was also relatively leachable, it was rejected from further consideration. The remaining three borosilicate frits (frits 111, 1806, and 1415) all yielded void-free melts at 950 °C, but shared a common characteristic: a second phase was formed similar to that observed with coke bottle glass. The most yellow phase was found in melts prepared with frit 1415. Frits 111 and 1806 were about equal in their tendency to form the yellow phase. Since frit 111 was much less leachable it was chosen for development studies.

Because the yellow phase has several undesirable features, discussed in more detail later, considerable effort was expended in determining methods of minimizing the amount of yellow phase present. The composition of the yellow phase is quite complex, but its presence in borosilicate-waste melts has been traced to one element, molybdenum, a high yield fission product. The yellow phase is not observed in either borate or phosphate-waste melts. Thus it can be concluded that silicate concentration in the melt is an important factor. Additions of phosphate to frit 111 prevented yellow phase formation but the melts were somewhat porous and the corrosion problem was increased. Additions of borate to frit 111 also decreased yellow phase formation although somewhat less effectively, and this was the solution finally adopted. Frit 111-15 represents a compromise between higher leach rates resulting from more borate and less yellow phase resulting from reduced silicate.

The allowable ratio of waste calcine to glass frit is very dependent upon processing temperature and frit particle size. The ratio can be increased at higher temperatures and with smaller frit size. For the WSEP runs the upper temperature limit is 950 °C and the frit size is -6+10 mesh. The need for the relatively large frit particles is imposed by the configuration of the WSEP solid flux addition system. The maximum allowable ratio of calcine to frit in the WSEP runs appears to be about 30:70 by weight. Laboratory tests indicate that this ratio could be 50:50 if -100 mesh frit were used. In laboratory studies of combining waste calcine and glass frit conducted at other sites -100 mesh frit was used.*

Various cations were added to the waste prior to calcination and their effectiveness in preventing formation of the molybdenum second phase was evaluated. At the suggestion of Dr. W. Guber of the Karlsruhe Research Center, titanium was included in the tests. Although large amounts were required (over one mole per liter in waste at 378 liters per tonne) titanium did inhibit separation of the yellow phase but mineralization in the product was increased. Several divalent cations also proved somewhat effective, and calcium and zinc at 0.65 and 0.50 moles per liter, respectively, were selected for use in Run SS-12. However, the optimum chemical technique for preventing formation of the molybdenum second phase in the in-pot melting flowsheet has not yet been found.

Results of laboratory tests have indicated that raising the operating temperature 100 °C, to 1050 °C, eliminates the

* *Bimonthly reports, Chemical Technology Group, Chemistry and Chemical Engineering Division, Nuclear Engineering Department, Brookhaven National Laboratory. July, 1965 through August, 1967.*

H. W. Godbee, N.M. Davis, Dispersion of Simulated High-Level Radioactive Waste Powders in a Glass Matrix, ORNL-TM-1897. August 4, 1967.

yellow phase completely. Gravimetric experiments show that since less than 5% of the yellow phase is volatilized in 24 hr at 1050 °C, dissolution in the borosilicate melt must occur at the higher temperatures. When the processing temperature is 950 °C there is also some suspension of calcine particles within the glassy matrix. These dissolve rapidly at 1050 °C.

The yellow phase from developmental spray calciner Run DSC-41 product was collected and analyzed. Globules of yellow phase were found at the top and the bottom of the filled pot in the following amounts:

Top yellow phase	Approximately 50 g.
Bottom yellow phase	Approximately 100 g.
Total weight of melt	Approximately 89.1 kg.

Some of the properties of the yellow phase are compared with the bulk phase in Table 5.3. Molybdenum is concentrated almost exclusively in the second phase. Of the long-lived fission products, strontium is not concentrated strongly in the yellow phase but cesium is. This was demonstrated in a special laboratory preparation (DSC-41 used potassium as a stand-in for cesium). The yellow phase contained 12.3 wt% cesium versus 2.5 wt% in the bulk glass.

Corrosion Studies - R. F. Maness

Corrosion of 304L and of 310 stainless steel was evaluated in a melt composed of 30 wt% PW-4m waste calcine from Run DSC-32* and 70 wt% of borosilicate glass frit 111-15 (see Table 5.2). Specimens of 304L exposed to the melt phase at 950, 1000 and 1050 °C corroded at rates of 0.02, 1.5, and 8.1 in./yr, respectively. Corrosion rates of specimens exposed to the melt-vapor interface were about 50% higher than those of

* BNWL-1186, 69-2.

TABLE 5.3. Characteristics of Second Phase in DSC-41 Product

	<u>Top Yellow Phase</u>	<u>Bulk Melt</u>	<u>Bottom Yellow Phase</u>
304L SS Corrosion rate at 950 °C, in./yr	0.44	0.02	0.44
Melting points, °C	700	950	975
Density, g/cm ³	3.116	3.102	3.731
Accelerated Leach Test at 950 °C			
% Weight Loss			
First 24 hr	78.2	1.4	22.8
Second 24 hr	4.4	1.1	1.1
Third 24 hr	3.8	1.1	0.68
Overall % wt loss	79.8	3.6	24.2
Composition, wt%(a)			
Nd	<1	0.8	1
Ce	<0.1	3	0.5
La	<0.1	3	1
Ba	6	3	5
Mo	7	0.3	8
Sr	0.8	3	5
Fe	1	1	0.05
Cr	1	0.1	0.2
Ca	1	20	10
K	6	0.7	5
Si	<1	8	<0.05
Na	60	30	20
B	0.07	3	0.4

a. The *analyses* were made by spark source mass spectrography and are estimated to be accurate to a factor of three.

specimens exposed to the melt phase only. Specimens of 310 exposed to the melt phase at 1000, 1050, and 1100 °C corroded at rates of 0.025, 0.19, and 2.1 in./yr, respectively. Corrosion rates of specimens exposed to the interface were about 508 higher, largely due to preferential attack at the interface rather than heavy vapor phase corrosion.

Analysis of Radioruthenium in WSEP Solidified Waste Products -
M. R. Schwab and F. E. Holt

A nondestructive direct count method was developed for the analysis of Ruthenium-106 in melt samples from the spray solidification and phosphate glass radioactive demonstration runs in WSEP. With the Direct Count method, the melt samples are weighed, then gamma scanned directly instead of dissolving the sample and gamma counting a small aqueous aliquot of the product sample. The gamma counting results for the dissolved product solutions have routinely given low ruthenium recoveries because of an incomplete dissolution of solid samples. Prior to Run PG-11, the total radioruthenium reported for each filled receiver pot was determined indirectly from the total Ruthenium-106 in the feed to the solidifier less the total Ruthenium-106 found in the condensates downstream from the solidifier. This indirect analysis indicated that from 80 to 95% of the input ruthenium should be contained in the phosphate glass solidified waste and from 25 to 50% should be contained in the spray solidified waste product. The results from the ruthenium balances for Runs PG-11, SS-11, and SS-12 are given in Table 5.4 with the Direct Count method used for the solid waste analyses. Good ruthenium recoveries are indicated. The Direct Count method for the first time actually verifies with a relatively high degree of confidence that the majority of the ruthenium not found downstream of the solidifier is indeed contained in the solidified waste.

**TABLE 5.4. Material Balances for Ruthenium-106 in WSEP
Radioactive Demonstrations (a)**

	Run PG-11	Run SS-11	Run SS-12
Product (b)	0.833	0.182	1.030
Evaporator (TK-113)	<0.005	0.73	0.019
Melter Condensate (TK- 117)	0.074	--	--
Total Recovery	0.91	0.91	1.05

- a. The data are reported as the fraction of Ruthenium-106 fed to the solidifier and the accuracy of data is within 5 to 10%.*
- b. Product analysis via the direct count method.*

ENGINEERING STUDIES

In-Pot Melting Development - J. D. Moore

Developmental spray solidifier Runs DSC-40 and 41 were made using the in-pot melting technique. During the runs simulated PW-4m waste with added calcium and zinc (see Table 5.5 for detailed feed compositions) was dried to a calcine in the spray calciner and the calcine was melted in the storage pot at 950 °C with a solid borosilicate glass frit. The zinc and calcium salts were added to improve melt product homogeneity. In DSC-41, zinc nitrate additive was used instead of zinc oxide (which was used in DSC-40) to reduce feed solids content to improve feed flow stability. With zinc nitrate, the feed solids content was only 10 vol% as compared to 40 vol% with zinc oxide. More stable feed flow was achieved with the lower solids-containing feed.

TABLE 5.5. PW-4m Feed Composition for Developmental Spray Solidifier Runs

Chemicals	Basis: 378 liters/tonne Concentration	
	Run Number	
	DSC-40	DSC-41
Fe^{+3}	0.128M ^(a)	0.128M ^(a)
Cr^{+3}	0.012	0.012
Ni^{+2}	0.051	0.051
Al^{+3}	0.001	0.001
Na^{+}	0.342 ^(b)	0.342 ^(b)
SiO_3^{-2}	0.01	0.01
PO_4^{-3}	0.003	0.003
NO_3^{-}	3.224	5.224
H^{+}	0.037	1.037
<u>Fission Products</u>		
MoO_4^{-2}	0.161	0.161
Sr^{+2}	0.036	0.036
Ba^{+2}	0.041	0.041
Rb + Cs (used K^{+})	0.092	0.092
(Y + RE) ⁺³	0.274	0.274
ZrO^{+2}	0.143	0.143
Ru^{+3}	0.0041 ^(c)	0.0041 ^(c)
Rh (used Co^{+2})	0.013	0.013
Ag^{+}	0.0016	0.0016
Cd^{+2}	0.0025	0.0025
Te (used SO_4^{-2})	0.014	0.014
Pd (used Ni^{+2} , included above)	0.043	0.043
Tc (used MoO_4^{-2} , included above)	0.031	0.031
<u>Additives</u>		
$\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$	0.65	0.65
ZnO	0.50	--
$\text{Zn}(\text{NO}_3)_2$	--	0.50
Glass frit ^(d) (added as solid to melt pot)	406 g/l (70% by weight of final product)	406 g/l

a. Nominal Fe concentration is 0.05M. Excess is Ru substitute.

b. Nominal Na concentration is 0.1M. Excess Na is from using $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$ for Mo addition.

c. Nominal Ru concentration is 0.082M.

d. 111-15, American Porcelain Enamel Co.

In DSC-40, borosilicate glass frit 111-15 (-6 mesh) was added to the pot at 5 min intervals. DSC-41 was run to determine if melt homogeneity would be affected if the frit addition frequency was decreased to once every 30 min. The once every 30 min addition was thought to be a practical frequency at which to add solids to the pot in the WSEP spray solidification process. In both runs, the average solids melting rate was 9.4 kg/hr including 6.6 kg/hr of glass frit. The larger frit batches used in DSC-41 taxed the pot melting capacity more than the smaller, more frequently added batches of DSC-40. In the latter run, the average temperature of the melt surface was higher (determined by brightness of color) than in DSC-41.

Both 304L stainless steel product pots were filled to a level of 30 to 33 in. with melt and maintained at a furnace temperature of 950 °C for 12 hr after the runs were shut down. After cooling, the pots were sectioned longitudinally for visual inspection. No evidence of pot wall or thermowell corrosion was found. The glass product of DSC-41 was not as homogeneous as the product from DSC-40. The product contained several voids up to 1 in. in diameter and the calcine did not appear to be as completely dissolved in the glass as it was in DSC-40. In the WSEP, however, the melt would be hotter in the center because of internal radioactive heat generation, and product homogeneity should be better than that observed in DSC-41. Also, increasing the solids addition frequency to at least once every 15 min should probably improve the product homogeneity.

Traces of a yellow crystalline material was interspersed throughout the product in both pots, but the material was much more abundant (about 1% of total melt volume) in the melt from DSC-41 as discussed in a previous section. The glassy melt in the bottom 4 in. of both pots had a mottled gray color, while the rest of the glass was dark brown. Although chemical

analyses of product samples showed that ruthenium was 4 to 9 times more concentrated in the bottom than in the top of the pot, the gray color could not be attributed to ruthenium since dark brown glass samples near the bottom of the pot contained about the same amount of ruthenium as the gray-colored samples.

Leach tests on glassy melt samples from both pots were conducted to determine if product solubility was affected by the flux addition frequency. Samples were leached for three days in distilled water heated to 95 °C. Water was changed every 24 hr, and the samples were weighed to determine weight loss from leaching. It was found that the product solubilities were not significantly different (3% soluble for DSC-40 and 3.9% soluble for DSC-41).

Spray calciner operation was relatively smooth in both runs. Operating conditions and results of both runs are summarized in Table 5.6. In DSC-40, feed flow was not as stable as in DSC-41 for reasons mentioned previously, and the spray nozzle plugged four times early in the run. The flow was maintained, however, when the atomizing air flow was reduced from 6.3 to 5.5 scfm. Calcine deposits on the inner walls of the calciner were less than 1/8-in. thick following both runs. Visual access through the glass window into the pot was adequate since it was necessary to see only the red glow from inside the pot. The inner side of the window was coated with a thin layer of powder, but vision was not fully blocked.

Less than 1% of the feed ruthenium was volatilized in DSC-40 and only 6% volatilized during DSC-41.

It was concluded from these runs that an adequate solid product could be attained by fusing PW-4m waste calcine with borosilicate frit 111-15 using the in-pot melting technique; this flowsheet was chosen for demonstration in WSEP.

TABLE 5.6. Summary of Developmental Spray Solidifier Runs

Run Number	DSC-40	DSC-41
Feed type	PW-4m	PW-4m
Feed concentration, liters/tonne	473	416
Additives	Glass frit (solid add to pot), $\text{Ca}(\text{NO}_3)_2$, ZnO	Glass frit (solid add to pot), $\text{Ca}(\text{NO}_3)_2$, $\text{Zn}(\text{NO}_3)_2$
Feed total acidity, M H^+ at 378 liters/tonne	0.53	2.57
Feed consumed, liters	205	150
Average feed rate, liters/hr	20.5	18
Calciner furnace temperature, °C	700	700
Calciner vacuum, inches of water	8	8
Atomizing air	air	air
Atomizing glass flow, scfm	5.5 to 6.3	5.7
Filter blowback pressure, psig	30(steam)	42(air)
Filter pressure drop, end of run, inches of water	7	4
Melt storage pot temperature, °C	950	950
Solid glass frit added to melt pot, kg	55.4	56.4
Frit addition frequency, min	5	30
Product collected, kg	79	84.1
Product density, g/cm^3	3.1	3.1
Product solubility, wt%(a)	3.0	3.9
Product concentration, liters/tonne	59	75
Particulate DF, feed to condensate	770	>4000
Ruthenium volatilized, % of total feed ruthenium	<1	6

a. *Product Leached for 3-day period in 95 °C distilled water. Water changed every 24 hr.*

RADIOACTIVE DEMONSTRATIONSSpray Solidification Run SS-11 - W. R. Bond

The twenty-eighth pilot plant demonstration of solidification of high level radioactive waste was successfully completed with the eleventh spray solidification Run SS-11. Run SS-11 was the first spray solidifier run with LMFBR waste; however, the waste as processed was not too different from the PW-4m waste that was demonstrated in Runs SS-9 and SS-10.* Differences were a slightly changed fission product spectrum and the addition of selenium, tellurium, antimony, and tin since these elements become more significant in the higher burnup fast reactor fuels. The completion of Run SS-11 marks the completion of the radioactive spray solidifier runs using a high temperature platinum melter.

During Run SS-11, a simulated waste equivalent to that resulting from the reprocessing of 0.54 tonne of LMFBR core fuel (100,000 MWd/tonne at 200 MW/tonne) with an equivalent aging time of 2.2 years was processed to produce 186 kg of ceramic product. The 60 liters of radioactive ceramic produced a heat rate density of 170 W/liter (10,900 W total based on analytical data and 10,200 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 759 °C, and the centerline-to-wall temperature difference was 353 °C. These values indicate an effective thermal conductivity of 1.24 W/(m²) (°C/m) [0.72 Btu/(hr) (ft²) (°F/ft)] for the ceramic product.

The solidifier was fed 669 liters of adjusted LMFBR feed (additives of 3.4M NaOH, 2.17M Fe(NO₃)₃, and 5.6M H₃PO₄ based on 378 liters/tonne) at 1230 liters/tonne in 45 hr. The feed containing 3,110,000 Ci of radioactivity including 14,400

* BNWL-1074, 69-1 and BNWL-1186, 69-2.

Ci of radioactive ruthenium was reduced in volume by a factor of 11.2, or 3.4 on a 378 liters/tonne basis. A summary of run data for Run SS-11 is presented in Table 5.7, and the feed composition for the run is given in Table 5.8.

TABLE 5.7. Summary Data for WSEP Spray Solidification Run SS-11

Operational Data

Date	3/2/70-3/4/70
Operating Mode	A
Feed Type	LMFBR
Feed, liters/tonne	1230
Total Feed to Solidifier, liters	669
Total Feed Time, hr	45
Average Feed Rate, liters/hr	14.9
Equivalent Waste to Solidifier, tonne	0.54
Volume Reduction, Feed Volume/Product Volume	11.2
Volume Reduction if Feed Were at 378 liters/tonne	3.4

Filled Pot Data

Total Curies in Feed to Pot	3,110,000
Total Curies of Ru in Feed to Solidifier	14,400
Product in Pot, liters	60
Product in Pot, liters/tonne	111
Heat Generation Rate in Pot, W	10,200
Equivalent Aging Time, years	2.2 ^(a)
£ Temperature in "Air-Cooled" Furnace, °C	759
£ to Wall Temperature Difference, °C	353
Effective Thermal Conductivity of Product in an Air-Cooled Furnace, W/(m ²) (°C/m)	1.24

a. Based on core fuel only at 100,000 MWd/tonne and 200 MW/tonne.

TABLE 5.8. LMFBF Feed Composition for Spray Solidification Run SS-11

<u>Component</u>	<u>Concentration, M^(a)</u>
H ⁺	---
Fe ⁺³	0.16
Cr ⁺³	0.046
Ni ⁺²	0.023
Al ⁺³	---
Na ⁺	0.10
PO ₄ ⁻³	0.06
NO ₃ ⁻	~5.5
SO ₃ ⁻²	---
MoO ₄ ⁻² + Tc as MoO ₄ ⁻²	0.30
Sr ⁺²	0.04
Ba ⁺²	0.068
Rb ⁺ + Cs ⁺ (added K ⁺)	0.229
(Y + RE) ⁺³	0.528
ZrO ⁺² + Nb as ZrO ⁺²	0.222
Ru ⁺⁴	0.0054
Ru ⁺⁴ (added Fe ⁺³)	0.22
Rh ⁺² (added Co ⁺²)	0.06
Pd ⁺² (added Ni ⁺²)	0.16
Ag ⁺² (added Cu ⁺²)	0.019
Cd ⁺² (added Cu ⁺²)	0.010
TeO ₄ ⁻³	0.039
SeO ₄ ⁻³	0.005
Sn ⁺⁴ + In ⁺³ as Sn ⁺⁴	0.011
Sb ⁺³	0.003
<u>Additive to Feed</u>	
Na ⁺	3.40
Fe ^{+3(b)}	1.71
Al ^{+3(b)}	0.46
PO ₄ ⁻³	5.58
ΣM ⁺ /P	2.5

a. Molarities are at 378 liters/tonne.

b. Nominal Fe additive is 2.17M, however excess Al in the waste from the processing plant was used as a substitute for Fe.

The operation of the spray solidification systems was very smooth. The principal process factors (atomization, filter operations, atomizing air and feed flow control) performed very satisfactorily. A newly installed atomizing nozzle with improved atomizing air pressure control produced the steadiest internal calciner temperatures and calcine fall to the melter that has been observed during the radioactive spray runs. An inspection of the inside of the solidifier after the run revealed that essentially no deposition of calcine occurred during the run.

During the run 73% of the ruthenium in the feed processed was volatilized from the solidifier. Although the ruthenium volatility was higher than in some previous runs, it was in general expected for the phosphoric-acid-containing feed. Only 0.013% of the nonvolatiles in the feed processed during the run were entrained from the solidifier. This represents a cumulative decontamination factor (DF) for nonvolatiles across the solidifier off-gas filters of 7.7×10^3 which is in agreement with instantaneous DF's which ranged from 2.9×10^3 to 6.0×10^4 .

The LMFBR melt was batch discharged from the melter via the drain freeze-valve into the unheated stainless steel receiver pot. The melter drain rate during discharge averaged approximately 34 liters/hr which is about a 40% faster drain rate than that attained with PW-4m melt during Run SS-10.* Although it was necessary to reduce the melter temperature approximately 50 to 100 °C after each dump to seal the freeze valve and stop the melter from dripping, an even fill of the receiver pot was achieved.

Following the run the platinum melter was removed from the melter furnace and it was found that the melter had four large indentations in the side of it as shown in Figure 5.1. The melter at the time of removal had experienced approximately 650 hr

* BNWL-1186, 69-2.

Neg 701124-2 Position I

Neg 701124-4 Position II

Neg 701124-5 Position III

FIGURE 5.1. Distorted Srray Solidifier High Temperature Platinum Melter

of intermittent service with temperatures ranging from 900 to 1200 °C and a vacuum of 0 to 20 in. of water, The melter had been visually examined prior to Run SS-5 after approximately 220 hr of service, and it had one slight indentation located in the same area as the indentation labeled number 2 in Figure 5.1, but it was considerably less pronounced. The exact cause of the distortion of the platinum melter is not known at this time, but is to be investigated further, A similar distortion of a platinum melter occurred at Brookhaven National Laboratory* and was attributed to the negative melter operating pressure and the reduced creep resistance of the platinum caused by the long process exposures, A subsequent Brookhaven melter was fabricated with increased wall thickness and stiffener rings welded to the outside melter wall to increase strength.

Auxiliary Process Equipment Performance During Run SS-11 -
J. N. Hartley

During Run SS-11 the WSEP auxiliaries were operated in a Mode A arrangement as illustrated in Figure 5.2. The radio-nuclide distribution for Run SS-11 is summarized in Table 5.9.

A modified solidifier condenser and condenser off-gas filtering system was installed prior to Run SS-11 to improve decontamination of the solidifier off-gas. The modified solidifier condenser and off-gas filtering system is shown in Figure 5.3. The bottom half of the condenser tubes were filled with pall rings (reduced from 5/8-in. to 1/2-in. diameter by mechanical press) to improve the condenser performance, Filling the tubes completely would have substantially increased the

* R. F. Drager, L. C. Emma, J. J. Fedele, L. P. Hatch, Gerald Strickland, E. J. Tuthill, and G. G. Weth. Development of the Phosphate Glass Process for Ultimate Disposal of High-Level Radioactive Wastes, BBL-50130(T-505). Brookhaven National Laboratory, Upton, New York, January 1968.

FIGURE 5.2, WSEP Auxiliary Process Equipment (Mod A Arrangement for Run SS-11)

Neg 700405-2

Neg 700405-5

FIGURE 5.3. New Solidifier Condenser and Offgas Filtering System

TABLE 5.9. Radionuclide Distribution in WSEP
Auxiliaries for Run SS-11

	Fraction of Equivalent Feed Radioactivity For Total Run in Effluent Streams	
	Ru-106	CePr-144
Evaporator (TK-113)	7.3×10^{-1}	1.3×10^{-4}
Fractionator (TK-115)	2.1×10^{-6}	$<4 \times 10^{-7}$
Fractionator Distillate Receiver (TK-116)	5.3×10^{-6}	8×10^{-10}
Scrubber (TK-118)	1.9×10^{-5}	7.7×10^{-9}
Offgas from Scrubber	1.8×10^{-8}	1.4×10^{-11}
Offgas to Stack	$<2 \times 10^{-10}$	3×10^{-12}
Ratio (Fractionator Distillate Radioactivity to 10CFR20)	1.6×10^4	2.2×10^2
Ratio (Scrubber Offgas Radio- activity to 10CFR20)	4.9×10^2	3.8×10^1
Ratio (Stack Offgas Radio- activity to 10CFR20)	$<4 \times 10^{-4}$	7.8×10^{-3}

pressure drop across the condenser to a point that vacuum requirements for the spray solidifier could not be met.

Radionuclides in the process off-gas are shown in Table 5.10. The solidifier condenser off-gas contained an average of $5.7 \times 10^{-3} \mu\text{Ci}/\text{cm}^3$ of Ru-106 and $4.3 \times 10^{-5} \mu\text{Ci}/\text{cm}^3$ of CePr-144. The cumulative decontamination factors (DF's) across the solidifier condenser were 1400 for Ru-106 and 6500 for CePr-144. This was an improvement over previous results by factors of 10 to 100. The off-gas leaving the solidifier condenser off-gas filter contained an average of $2.8 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ of Ru-106 and $2.6 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ of CePr-144. These values indicated average instantaneous DF's across the filter of 2.0×10^3 for Ru-106 and 16 for CePr-144.

The accumulation of radioruthenium in the auxiliaries as seen in Figure 5.4 was generally typical of previous runs. However, the radioruthenium accumulation in the fractionator was not linear as was the case in previous runs, The ruthenium

TABLE 5.10. Radionuclides in Process Offgas

Sample Number	Sample Period		Solidifier Condenser Offgas, $\mu\text{Ci}/\text{cm}^3$		Offgas from Solidifier Condenser Filter, $\mu\text{Ci}/\text{cm}^3$		Evaporator Condenser Offgas, $\mu\text{Ci}/\text{cm}^3$	
	Start	End	Ru-106	CePr-144	Ru-106	CePr-144	Ru-106	CePr-144
1	6.5	9.8	2.9×10^{-5}	1.5×10^{-5}	1.1×10^{-6}	5.6×10^{-6}	1.1×10^{-7}	4.3×10^{-7}
2	14.0	17.3	1.0×10^{-2}	1.1×10^{-4}	2.0×10^{-6}	1.3×10^{-6}	6.2×10^{-7}	1.5×10^{-7}
3	20.2	23.3	7.5×10^{-5}	3.2×10^{-5}	1.9×10^{-7}	7.5×10^{-7}	3.8×10^{-7}	1.4×10^{-6}
4	31.8	34.8	9.2×10^{-4}	2.1×10^{-5}	---	---	2.6×10^{-7}	6.1×10^{-7}
5	38.3	41.0	7.1×10^{-3}	3.6×10^{-5}	7.5×10^{-6}	2.8×10^{-6}	3.7×10^{-7}	2.2×10^{-6}
6	52.0	55.0	---	---	3.4×10^{-6}	2.5×10^{-6}	2.9×10^{-7}	7.8×10^{-7}
	Average		5.7×10^{-5}	4.3×10^{-5}	2.8×10^{-6}	2.6×10^{-6}	3.4×10^{-7}	9.3×10^{-7}

Sample Number	Sample Period		Fractionator Condenser Offgas, $\mu\text{Ci}/\text{cm}^3$		Offgas from F-112 Absolute Filter, $\mu\text{Ci}/\text{cm}^3$		Final Offgas from Scrubber			
	Start	End	Ru-106	CePr-144	Ru-106	CePr-144	$\mu\text{Ci}/\text{cm}^3$		Ratio to 10CFR20	
							Ru-106	CePr-144	Ru-106	CePr-144
1	-5.8	-1.3	$3.4 \times 10^{-8(a)}$	1.1×10^{-7}	5.8×10^{-9}	1.4×10^{-8}	1.0×10^{-10}	4.1×10^{-9}	5×10^{-1}	1.0×10^1
2	0.3	8.5	3.2×10^{-6}	1.6×10^{-7}	8.4×10^{-8}	---	6.1×10^{-8}	2.5×10^{-8}	3.1×10^2	6.3×10^1
3	11.0	16.0	9.5×10^{-6}	2.7×10^{-7}	$1.1 \times 10^{-7(a)}$	---	6.7×10^{-8}	---	3.4×10^2	---
4	16.5	24.5	6.0×10^{-6}	9.3×10^{-8}	4.9×10^{-9}	1.2×10^{-8}	1.5×10^{-8}	---	7.5×10^1	---
5	25.0	31.8	5.8×10^{-6}	9.9×10^{-7}	4.6×10^{-9}	---	4.1×10^{-8}	---	2.0×10^2	----
6	32.8	35.0	8.3×10^{-6}	3.4×10^{-7}	5.5×10^{-9}	---	1.0×10^{-9}	---	5.0	----
7	35.0	45.5	5.2×10^{-6}	2.7×10^{-7}	5.5×10^{-9}	---	2.0×10^{-10}	---	1.0	---
8	45.5	54.5	2.2×10^{-6}	4.3×10^{-8}	9.8×10^{-8}	---	5.0×10^{-9}	---	2.5×10^1	---
	Average		5.0×10^{-6}	3.3×10^{-7}	4.4×10^{-8}	1.3×10^{-8}	9.8×10^{-8}	1.5×10^{-8}	4.9×10^2	3.8×10^1

a. Excluded from averages

FIGURE 5.4. Radioruthenium Accumulation in WSEP Auxiliaries During Run SS-11

accumulation seemed to level out after an initial jump early in the run. Basing the accumulation in the fractionator on the evaporator condensate stream samples, the total accumulation should have been a factor of about 20 less. This fact indicates that the ruthenium accumulation may have been caused primarily by internal contamination early in the run.

The auxiliary evaporator operated on about 5.7M HNO_3 with an overhead acidity ranging from 0.1 to 0.8M HNO_3 . Instantaneous ruthenium DF's across the evaporator ranged from 4.4×10^3 to 1.3×10^5 . These values were generally a factor of 13 to 100 greater than in previous runs with feed on to the solidifier, indicating (along with gas sample results) that the new modified condenser and off-gas filter were stopping the carryover of ruthenium in the noncondensable gas stream from the solidifier condenser. However, there was again no real apparent correlation between evaporator overhead acidity and ruthenium instantaneous DF's. This was attributed to oxides of nitrogen not being scrubbed out completely by the solidifier condenser. The solidifier condenser only stopped 41% of the total nitrogen in the solidifier off-gas. Part of the remaining 59% was scrubbed out by the evaporator condenser increasing the evaporator overhead acidity, and resulting in no correlation between ruthenium instantaneous DF's and overhead acidity. That ruthenium equivalent to $2.1 \times 10^{-1}\%$ of that fed to the solidifier accumulated in the fractionator indicated a cumulative ruthenium DF of 350 across the evaporator, and that less than $4 \times 10^{-5}\%$ of the CePr-144 representing nonvolatiles accumulated in the fractionator indicated a cumulative DF for nonvolatiles of greater than 4×10^2 .

The cumulative DF across the fractionator was 6.3×10^2 for Ru-106 and 1.3×10^3 for CePr-144 representing nonvolatiles.

During Run SS-11, 80% of the fractionator distillate was recycled to the evaporator as strip water while the remaining 20% was accumulated in the distillate receiver. The fraction of equivalent feed radioactivity that accumulated in the fractionator distillate receiver was 5.3×10^{-6} for Ru-106 and 8×10^{-10} for CePr-144. The concentration of radionuclide in the accumulated fractionator distillate (final aqueous effluent) was above 10CFR20 release limits by 1.6×10^4 for Ru-106 and 2.2×10^2 for CePr-144.

The off-gas leaving the fractionator condenser contained an average of $5 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ of Ru-106 and $3.3 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$ CePr-144. The off-gas is filtered through a high efficiency filter before entering the scrubber. The off-gas leaving the filter contained $4.4 \times 10^{-8} \mu\text{Ci}/\text{cm}^3$ of Ru-106 and $1.3 \times 10^{-8} \mu\text{Ci}/\text{cm}^3$ CePr-144 which indicated DF's across the filter of 1.1×10^2 for Ru-106 and 25 for CePr-144 (representing particulate DF). The DF for particulates was lower by a factor of about 100 than in previous runs.

The fraction of equivalent feed radioactivity that accumulated in the off-gas scrubber was 1.8×10^{-8} for Ru-106 and 1.4×10^{-11} for CePr-144. However, using the results from gas samples taken prior to the scrubber, the fraction of feed radioactivity was 3.8×10^{-9} for Ru-106 and 5.8×10^{-12} for CePr-144. This discrepancy could have been caused by internal contamination in the scrubber and piping leading to the scrubber.

The off-gas scrubber was inadvertently started with 0.44M NaOH instead of the usual 2M NaOH. The scrubber became neutralized after 30% of the feed had been fed to the solidifier at which time the accumulation rate of nitrogen dropped off rapidly until at about 60% feed fed no more nitrogen accumulated. Figure 5.5 shows data from the auxiliary scrubber operation. A total of 15% of the nitrogen fed to the solidifier accumulated in the scrubber. Using the initial nitrogen accumulation rate

FIGURE 5.5. Auxiliary Scrubber Operation

before the scrubber became neutralized and extrapolating for the entire run indicates that about 30% of the feed nitrogen should have accumulated in the scrubber. Therefore, about 15% escaped with the scrubber off-gas.

The final process off-gas leaving the scrubber contained an average of $9.8 \times 10^{-8} \mu\text{Ci}/\text{cm}^3$ of Ru-106 and $1.5 \times 10^{-8} \mu\text{Ci}/\text{cm}^3$ of CePr-144. These values were above 10CFR20 release limits by factors of 4.9×10^2 for Ru-106 and 3.8×10^1 for CePr-144. However, after the scrubber off-gas was filtered twice more and combined with building ventilation air, the radioactivity released with the off-gas to the stack was well below 10CFR20 release limits.

Spray Solidification Run SS-12(IPM) - W. R. Bond

Run SS-12(IPM) was successfully completed using the in-pot melting technique wherein spray calcined PW-4m waste was melted with a borosilicate glass frit directly in the final storage container. The WSEP spray calciner had been coupled directly to the receiver pot for this run via a special spool-piece, and the solid frit was added directly to the pot via a solids feeder attached to the spool piece.

During Run SS-12(IPM), a simulated waste equivalent to that resulting from the reprocessing of 0.54 tonne of power reactor fuel (45,000 MWd/tonne at 30 MW/tonne) with an equivalent aging time of 1.3 years was processed to produce 162 kg of product. The 54 liters of radioactive solid produced a heat rate density of 89 W/liter (4900 W total based on analytical data and 4800 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 474 °C, and the centerline-to-wall temperature difference was 197 °C. These values indicate an effective thermal conductivity of $1.12 \text{ W/(m}^2)(^{\circ}\text{C/m)}$ [$0.65 \text{ Btu/(hr)(ft}^2)(^{\circ}\text{F/ft)}$] for the product. The effective thermal conductivity of the product while molten during processing was approximately 3 to 4 times higher than this and consequently pot centerline temperatures did not exceed 980 °C despite pot wall temperatures of 900 to 950 °C.

The calciner was fed 301 liters of adjusted PW-4m feed (additives of 0.65M $\text{Ca}(\text{NO}_3)_2$ and 0.5M $\text{Zn}(\text{NO}_3)_2$ based on 378 liters/tonne) at 556 liters/tonne in 28 hr. The resulting calcine was combined with 109 kg of borosilicate glass frit in the receiver pot, The feed containing 1,380,000 Ci of radioactivity including 6800 Ci of radioactive ruthenium was reduced in volume by a factor of 5.6, or 3.8 on a 378 liters/tonne basis, A summary of run data for SS-12 is presented in Table 5.11 and the feed composition for the run is given in Table 5.12.

TABLE 5.11. Summary Data for WSEP Spray Solidification
Run SS-12(IPM)

Operational Data

Date	4/6/70-4/7/70
Operating Mode	A
Feed Type	PW-4m
Feed, liters/tonne	556
Total Feed to Solidifier, liters	301
Total Feed Time, hr	28
Average Feed Rate, liters/hr	10.8
Equivalent Waste To Solidifier, tonne	0.54
Volume Reduction, Feed Volume/Product Volume	5.6
Volume Reduction if Feed were at 378 liters/tonne	3.8

Filled Pot Data

Total Curies in Feed to Pot	1,380,000
Total Curies of Ru in Feed to Solidifier	6800
Product in Pot, liters	54
Product in Pot, liters/tonne	100 ^(a)
Heat Generation Rate in Pot, W	4800
Equivalent Aging Time, years	1.3
Core Temperature in "Air-Cooled" Furnace, °C	474
Core to Wall Temperature Difference, °C	197
Effective Thermal Conductivity of Product in an Air-Cooled Furnace, W/(m ²)(°C/m)	1.12

a. This is 15% higher than desired, due to the addition of extra borosilicate flux to compensate for excess sodium and aluminum in the feed.

TABLE 5.12. **PW-4m Feed Composition for Spray Solidification
Run SS-12(IPM)**

<u>Component</u>	<u>Concentration, M^(a)</u>
H ⁺	---
Fe ⁺³	0.05
Cr ⁺³	0.012
Ni ⁺²	0.008
Al ⁺³	0.287 ^(b)
Na ⁺	0.816 ^(c)
PO ₄ ⁻³	0.003
NO ₃ ⁻	~2.6 ^(d)
Sr	---
MoO ₄ ⁻² + Tc as MoO ₄ ⁻²	0.161
Sr ⁺²	0.036
Ba ⁺²	0.041
Rb ⁺ + Cs ⁺ (added K ⁺)	0.092
(Y + RE) ⁺³	0.325 ^(e)
ZrO ⁺² + Nb as ZrO ⁺²	0.143
Ru ⁺⁴	0.0041
Ru ⁺⁴ (added Al ⁺³)	0.078
Rh ⁺² (added Co ⁺²)	0.013
Pd ⁺² (added Ni ⁺²)	0.043
Ag ⁺² (added Cu ⁺²)	0.0016
Cd ⁺² (added Cu ⁺²)	0.0025
<u>Additive to Feed</u>	
Ca ⁺²	0.65
Zn ⁺²	0.50
<u>Additive to Pot</u>	
Borosilicate glass frit	537 g/liter ^(f)

- a. Molarities are at 378 liters/tonne.
b. The nominal Al concentration is 0.001M, however excess Al was present in the waste from the processing plant used to simulate the PW-4m feed.
c. The nominal Na concentration is 0.1M, however 0.322M Na was added as Na₂MoO₄ and the remaining excess resulted from the processing plant waste.
d. Does not include the 2.3M NO₃⁻ added with the additives.
e. The nominal RE concentration is 0.274M, however excess RE was present in the processing plant waste.
f. For the nominal PW-4M feed the borosilicate frit requirement is 478 g/liter, however extra frit was added to compensate for excess Na and Al in the feed.

During the first 4 hr of Run SS-12(IPM) the spray calciner average feed rate was approximately 15 liters/hr, and the in-pot melting process easily kept up with the calciner at a melt forming rate of approximately 2.7 liters/hr. The normal melt forming capacity for an 8-in. diameter pot is expected to exceed 3 liters/hr based on cold development runs or more than double the melt forming capacity of the 10-in. diameter spray and phosphate glass melters. During this run however, the spray calciner capacity was limited for some unknown reason and except for the first few hours of the run it was not possible to exceed a feed rate of 11 liters/hr (a melt forming rate of 2.0 liters/hr) without producing a significant drop in the calciner internal temperatures. Although poor feed atomization would be suspected as a cause for the reduced capacity, the atomizing air and feed flow control were very satisfactory during the run. An inspection of the feed nozzle and the internals of the calciner is planned to determine the cause of the problem. A decrease in calciner capacity did not occur with the same type feed during cold development spray calcine runs.

The spray calciner feed was interrupted twice during Run SS-12(IPM) for a total of 4.4 hr. The first shutdown was made to enable an evaluation of why the calciner capacity was limited, and the second was made while a failed calciner vibrator was replaced. The performance of the spray calciner off-gas filters was very satisfactory during the run as the filter pressure drop was maintained at 3.3 in. of water and only 0.014% of the particulates were entrained from the calciner.

Ruthenium volatilization from the solidifier during SS-12(IPM) was only 1.9% of the ruthenium in the feed to the solidifier. This compares with previous spray runs where

40 to 75% of the ruthenium was volatilized, The decrease during this run was anticipated from the results of cold development runs and is explained by the absence of phosphoric acid in the feed to the spray calciner,

HIGH-LEVEL WASTES FROM ADVANCED REPROCESSING TECHNIQUES

K. J. Schneider

A review of advancements in fuel reprocessing technology was made to determine the effects of likely changes on the compositions of high-level wastes in the Waste Solidification Demonstration Program. It was concluded that all known near-future changes in waste compositions have been included in the existing waste program. Wastes from aqueous reprocessing of LWR fuels are bracketed by the reference WSEP compositions PW-1, PW-2, and PW-4m. Wastes from aqueous reprocessing of LMFBR fuels are bracketed by the above reference composition plus the reference LMFBR composition. For either LWR or LMFBR fuels non-aqueous reprocessing techniques which significantly change the waste composition are not yet well enough defined to do major studies on the high-level waste. A discussion of the anticipated changes in high-level wastes from reprocessing of LMFBR fuels is presented below,

The higher fuel exposure and specific power and the use of plutonium enrichment rather than Uranium-235 in the core fuel of LMFBR's will result in higher fission product contents and heat generation rates, and in a different distribution of fission products in the waste. These differences will be highly dependent upon the reactor design, and ultimately upon future decisions relating to the processing of blanket fuel with or without core fuel. The importance of these items can be best illustrated by Table 5-13 which presents conceptual fast reactor characteristics,

TABLE 5.13. Summary of Conceptual LMFBR Characteristics (a)

LMFBR Type	Equilibrium Burnup, MWd/MT				Equilibrium Refueling Rate, MT/yr for each 1000 MW _e Reactor Producing 310,000 MW _e /yr		
	Core	Axial Blanket	Radial Blanket	Average ^(b)	Core	Axial Blanket	Radial Blanket
Reference Oxide	80,000	2,500	8,100	33,000	8.497	4.952	10.04
Advanced Oxide ^(c)	96,500	2,500	6,000	35,600	6.764	5.535	7.845
Advanced Oxide ^(d)	97,500	2,800	5,700	26,600	6.289	5.893	14.659
Advanced Oxide ^(e)	104,500	8,700	9,000	41,700	6.131	6.884	4.798
Carbide	79,500	3,000	3,800	19,600	8.973	5.611	3.169
Advanced Carbide	110,300	5,900	8,300	47,300	6.455	2.670	7.591

- a. Data from WASH-1099, "Reactor Fuel Cycle Costs for Nuclear Power Evaluation", (Report of the Fuel Recycle Task Force), Draft II.*
- b. Weighted average of core, axial blanket and radial blanket.*
- c. Positive sodium coefficient.*
- d. Negative sodium coefficient.*
- e. Data from General Electric Company, "Fast Breeder Reactor Follow-on Study", 1969.*

The equilibrium burnup (in terms of the weighted average of the core fuel, axial blankets, and radial blanket) is in the range of projected thermal reactor fuel in most cases. However, this equilibrium burnup is based upon the assumption that the radial blanket will be mixed with the core and axial blanket during fuel processing. This mode of operation has not yet been demonstrated to be the most economical. It may be more economical to process the radial blanket separately in some other manner. In most cases, the core and axial blankets will be contained in the same fuel pin and will be blended during processing. If the radial blanket is processed separately, the equilibrium burnup will be much higher for the mixture of core fuel and axial blanket.

It must also be stressed that the heat generation rate will be higher (especially at short cooling times) in a mixture of fast reactor fuel with a given equilibrium or average burnup than in a thermal reactor fuel with the same burnup. This difference occurs because, in the case of the mixed fast reactor fuel, most of the fission product radioactivity is contributed by the core fuel. The core fuel operates at a much higher specific power than thermal reactor fuel and the production rate and concentration of short-lived fission products increases as the specific power increases.

The fission product spectrum will also be different in waste from fast reactor fuel processing. This difference is due to the different irradiation conditions, the plutonium enrichment in the fuel, and higher neutron energy in a fast reactor. For example, the relative amounts of Ruthenium-106 will be increased several-fold in wastes from processing of fast reactor fuel. The relative amounts of cadmium, tin, antimony, selenium, and tellurium in the core fuel will also be increased by factors

up to 3, Some of these latter fission products, notably selenium, tellurium, and antimony, tend to be volatile like ruthenium at high temperatures and could complicate waste solidification processes.

Table 5.13 shows that the burnup of the core fuel in the conceptual "advanced" reactors is higher than in the reference reactors. Thus, the higher burnups, the higher specific power in fast reactor core fuel, the different spectrum of fission products in the fuel, and the possibility that the radial blanket will be processed separately require evaluation in solidification studies with fast reactor wastes.

The future potential of carbide-fueled LMFBRs is not clear at this time. Follow-on studies of LMFBR conceptual designs have indicated anticipated improved performance of oxide-fueled reactors compared to carbide-fueled reactors. Consequently, studies with high-level wastes from carbide-fueled reactors are believed to be unnecessary until the carbide-fueled reactors appear more promising.

In the Waste Solidification Demonstration Program, the total fission product content of wastes from LMFBRs, radioactive plus nonradioactive, was selected to be representative of a core fuel irradiated to 100,000 MWd/tonne at 200 MW/tonne. A chemically clean waste composition was selected because such a composition represents the "worst" case in terms of storage temperatures, fission product concentration, and the amount of potentially volatile fission products per container. The other reference waste compositions in the Waste Solidification Demonstration Program are believed to bracket the other characteristics of most currently anticipated high-level waste compositions from LMFBR's.

Potential key nonradioactive constituents in high-level wastes from reprocessing of LMFBR fuels that may be present in large quantities are iron, (primarily from partial dissolution of the stainless steel cladding or from the use of ferrous ion as a chemical reducing agent) and sodium (primarily from treatment of the organic solvent). The probable maximum amounts of these constituents in combined wastes are shown in Table 5-14 and are compared with adjusted LMFBR waste compositions in WSEP demonstrations,

As shown, the concentration of the key constituents in the WSEP demonstrations with LMFBR wastes, in combination with the three reference LWR waste compositions used in the WSEP program, bracket the anticipated range of high-level waste compositions, even when combined with other fuel reprocessing wastes. Investigation of the other primary differences in wastes from LMFBR fuel reprocessing (namely the higher content of fission products, the larger amounts of potentially volatile fission products, and the higher levels of heat from fission product) have been included in the program studies.

EVALUATION OF SOLIDIFIED WASTE PRODUCTS

NONRADIOACTIVE LABORATORY STUDIES

Melt Segregation Studies of Simulated Solidified Wastes Stored at High Temperatures for Extended Periods - C. Hampton

A study of the behavior of simulated WSEP melts when stored at temperatures up to 950 °C was conducted. Simulated phosphate glass and spray solidifier PW-1, PW-2, PW-4m, and LMFBR melts were used in the study.

The solidified wastes were all melted at 950 °C to facilitate ease in handling. The resulting melts were then broken into pieces. A weighed amount of melt was placed in an alumina crucible and the weight of the entire assembly recorded. The crucible and sample were then placed in the storage furnace and

TABLE 5.14. Key Constituents in High-Level Wastes from Reprocessing of LMFBR Fuels

Constituent	Hypothetical Concentration at 100 gal Waste/tonne, M							
	Dissolution of 10% Cladding	Alpha (a) Waste Maximum	Solvent (a), (b) Cleanup Waste, Maximum	Counter (a) Scrubber Waste, Maximum	Maximum (a) Total	Probable (c) Minimum Total	Maximum Total in WSEP Spray Kuns	Total in WSEP Phosphate Glass Runs
Fe ²⁺	1.22	0.28	----	----	1.50	0.1	2.26	0.93
Na ⁺	----	----	1.02	0.44	1.46	0.1	2.96	3.06
Fission Products	----	----	----	----	1.4	----	4.1	4.1

a. The amounts shown obtained from R. E. Blanco, Oak Ridge National Laboratory, January, 1970.

b. Assumes one cycle using amine solvent.

c. The amounts shown obtained from G. L. Richardson, Battelle-Northwest, November, 1969.

heated to 950 °C and processed for approximately 1 hr. The final temperature was then adjusted to the storage temperature indicated in Table 5.15. When storage temperature was reached (within one day) the melts were maintained at this temperature for 8 weeks. At the end of 8 weeks the samples were cooled slowly over a 2 to 3 day period by slowly decreasing the furnace temperature. After cooling, the crucibles were sawed in half and the melts inspected visually for phase segregation and other physical changes. Later the samples were analyzed for elemental segregation by comparing composition of top and bottom phases. The solubility of top and bottom phases were also checked,

The thermal stability of phosphate glasses, with relationship to phase separation, is superior to that of spray melts. However, indications are that for long-term storage the end result is the same. The melts all showed phase separation or segregation at 950 °C after 8 weeks storage, comparable to that shown in Figure 5.6.

Spray solidifier melts at the lower temperatures of 750 °C and 850 °C appeared to be uniform or homogeneous. However, the phosphate glass melt; showed phase separation at 850 °C. No chemical analysis as yet has been made on the melts stored at these lower temperatures,

The phosphate glass PW-4m and spray solidifier PW-4m melts stored at 950 °C were chosen to represent phosphate glass and spray products that had been processed at 950 °C for 8 weeks. These samples were analyzed for cesium, ruthenium, strontium, and rare earths. Samples were taken from the top and bottom layer of each melt to compare elemental composition. The following results were obtained and are compared with the calculated results from the original elemental input as shown in Table 5.16.

TABLE 5.15. Melt Storage Temperature and Melt Condition After 8 Weeks

<u>Phosphate Glass Melts</u>		<u>850 °C</u>	<u>950 °C</u>	
PW-1		PS	PS	
PW-2		FS	FS	
PW-4m		FS	FS	
LMFBR		FS	FS	
<u>Spray Solidifier Melts</u>		<u>750 °C</u>	<u>850 °C</u>	<u>950 °C</u>
PW-1	H	H		PS
PW-2	H	H		PS
PW-4m	H	H		PS
LMFBR	H	H		PS

H - Homogeneous.

FS - Faint phase separation.

PS - Phase separation very distinct.

TABLE 5.16. Chemical Analysis of Phosphate Glass and Spray Solidifier Melts Stored at 950 °C for 8 Weeks

<u>Element</u>	<u>Theoretical wt% of Melt</u>			
	<u>Phosphate Glass</u>		<u>Spray Solidifier</u>	
	<u>PW-4m Melt</u>		<u>PW-4m Melt</u>	
Cs	2.06		2.65	
Ru	0.63		0.81	
PO ₄	87.2		62.0	
Sr	0.62		0.79	
RE	11.45		8.93	

<u>Element</u>	<u>Results of Analysis as wt%^(a)</u>			
	<u>Phosphate Glass PW-4m Melt</u>		<u>Spray Solidifier PW-4m Melt</u>	
	<u>Top Phase</u>	<u>Bottom Phase</u>	<u>Top Phase</u>	<u>Bottom Phase</u>
Cs	2.2	1.97	2.75	1.50
Ru	0.05	0.14	0.06	0.97
PO ₄	79.8	80.2	51.3	48.7
Sr	0.29	0.19	0.08	0.30
RE	7.0	9.95	2.85	8.0

as oxide

a. It should be noted that dissolving the products for analysis was a very difficult process and incomplete solution may account for some of the discrepancies between theoretical and actual weight percents.

Phosphate Glass Melt Stored at 950 °C

Spray Solidifier Melt Stored at 950 °C

FIGURE 5.6, Typical Phase Separation of Phosphate Glass and Spray Solidifier Melts Stored at 950 °C

Leachability tests were also run on the phases and are reported in Table 5.17.

TABLE 5.17. Leachability of Phosphate Glass and Spray Solidifier PW-Pm Melts Stored at 950 °C for 8 Weeks

<u>Spray Solidifier Melt</u>	<u>First 24 hr</u>	<u>Second 24 hr</u>	<u>Third 24 hr</u>	<u>Total</u>
Top Phase	10.29%	0.59%	0.19%	11.07%
Bottom Phase	8.22%	0.37%	0.10%	8.69%
<u>Phosphate Glass Melt</u>				
Top Phase	0.35%	0.59%	0.93%	1.87%
Bottom Phase	0.62%	1.41%	1.61%	3.64%

PRODUCT MEASUREMENT, TESTING AND STORAGE

Solids Storage Engineering Test Facility (SSETF)

R. J, Thompson

Table 5.18 shows the solidified waste containers that have been transferred to SSETF for environmental testing. The waste container from WSEP Run SS-10 was added to the environmental test program during this report period. The maximum container temperatures reported in Table 5.18 are somewhat lower than previously reported due to a minor interruption in electrical power to externally heated pots. The reduced temperatures are a short-time event and temperatures will be restored to the higher levels.

The irregular temperature profiles reported previously for WSEP Run SS-8 waste container has been resolved. The pot thermocouples were replaced and the irregular temperature readings eliminated. While the waste container was out of the environmental test pod, careful single thermocouple temperature scans were made of the intermediate and centerline thermowells. These scans substantiated the fact that temperature profile irregularities were due to faulty thermocouples.

TABLE 5.18. SSETF Environmental Test Summary (a)

<u>Run Number</u> <u>Pot Number</u> <u>Cubicle Number</u>	<u>Date</u> <u>Testing</u> <u>Started</u>	<u>Pod</u> <u>Environment</u>	<u>Maximum</u> <u>Temperature °C</u>		
			<u>Pod</u>	<u>Pot</u> <u>Wall</u>	<u>℄</u>
SS-4 40 A-21-3	9/11/69	Water	74	76	138
PG-6 39 A-21-1	10/1/69	Water	79	100	157
PC-6 30 A-21-4	11/17/69	Air	100	---	253
SS-8 43 A-21-2	1/5/70	Air	221	---	357
SS-10 54 A-31-3	2/9/70	Water	24	29	176

a. Temperature data recorded 4-14-70.

6. HEAVY SECTION STEEL TECHNOLOGY PROGRAM

IRRADIATION EFFECT ON THE FRACTURE OF HEAVY SECTION PRESSURE

VESSEL STEELS - C. W. Hunter, J. A. Williams,
and C. L. Hellerich

Specimen Irradiations

The irradiation facilities in use or under development are the ETR M-3 hot water loop for irradiation of 1-in.-thick fracture mechanics specimens (1T CT) and the ATR capsule for 4T CT specimens. The irradiation of specimens in the ETR M-3 loop during Cycles 101 through 104B have been previously described.⁽¹⁻³⁾ The testing of specimens from Cycle 101 is complete, as is the testing of tensile specimens from Cycle 102. Fatigue pre-cracks are being prepared in the 1T CT specimens from Cycles 102 and 103. The tensile and fracture specimens from 104B are being readied for testing. Irradiations in the M-3 loop will be commenced again with Cycle 108 in June; the anticipated cycles through February 1971 are 108, 109A, 111, and 112. Both transverse (WR) orientation base plate and weldment will be irradiated. Several of the six tubes containing tensile specimens will be modified to accept Charpy specimens.

The ATR - 4T irradiation facility will occupy 10 reflector positions ON-3 through ON-12. Specific studies of the fast neutron flux and gamma heating in these positions have been conducted recently. The gamma heating values are listed in Table 6.1. Positions ON-3, ON-4, and ON-5 are at the front face and hottest side of the specimen, while ON-9 is at the back face. These values were measured with graphite at a reactor power of 1.65 MW and extrapolated to 250 MW. These present values are deemed more accurate than the values previously reported.⁽³⁾ These previous values were from supposedly similar, but apparently not identical south reflector positions numbered OS-3 through OS-12. These latest values

indicate that the average gamma heating will be 0.30 W/g at the front face of the specimens and 0.15 at the back face. The analyses of the fast neutron flux in Position ON-3 through ON-12 have not yet been completed.

TABLE 6.1. Gamma Heat Generation ^(a) in Select Reflector Positions to be Occupied by the 4T Capsule in the ATR

Position Above Core Bottom, in.	ON-3 <u>W/g</u>	ON-4 <u>W/g</u>	ON-5 <u>W/g</u>	ON-9 <u>W/g</u>
9	0.21	0.21	0.29	0.11
15	0.26	0.29	0.34	0.14
21	0.26	0.33	0.39	0.16
27	0.26	0.32	0.39	0.16
33	0.24	0.27	0.34	0.14
39	0.19	0.23	0.26	0.12
45	0.12	0.15	0.18	0.08
51	0.07	0.08	0.08	0.05

a. Determined with graphite at 1.65 MW, Extrapolated to 250 MW.

The general features of the ATR - 4T capsule have been previously described.⁽³⁾ Two independent subcapsules with two specimens each will be employed. The four specimens will consist of two transverse (WR) orientation and one longitudinal (RW) orientation specimens of base material and one weld-metal specimen. The most critical feature of the design will be to maintain the necessary gas gap of approximately 0.012-in. between the specimens and the walls of the capsule, while maintaining the capability to withstand a hydrostatic force of 70,000 lb on the larger faces of the capsule. To meet this requirement, the inside walls of the capsule will be relief machined leaving small bearing pads distributed uniformly. In addition to maintaining the desired gap spacing, the bearing pads will remove about two-thirds of the heat generated. Irradiation is scheduled to commence ATR Cycle 5.

Base Material Studies of ASIM A533-B

The material being studied is quarter through center thickness locations from the 12-in. ASIM A533 Grade B Class I HSST Program Plate 02. Past and current efforts are centered with the fracture of RW orientation 1T CT and tensile specimens irradiated from 1 to 8×10^{19} n/cm² ($E > 1$ MeV) at 540 °F. Future efforts will be at the same irradiation conditions but will also include Charpy impact fracture and 4T CT specimens, and will emphasize the WR orientation behavior. The longitudinal tensile specimens irradiated at 2×10^{19} and 4.4×10^{19} n/cm² ($E > 1$ MeV) have been tested, as have some of the RW orientation 1T CT specimens at 2×10^{19} n/cm². Eight similarly irradiated 1T CT specimens are being fatigue pre-cracked. Testing emphasis with these will be to define more clearly both the low- and high-temperature portions of the K_{IC} curve.

The previously reported⁽³⁾ fluence effect tensile results has been replotted in Figure 6.1 to indicate the percentage increase in properties as a function of fluence. The room temperature properties are more responsive to increased fluence. Also the relative increase in yield strength is greater than that of ultimate strength and it is also more responsive to increased fluence. This behavior reduces the margin between yield and rupture.

The center plane of the 4T CT specimens will be 3.5 in. from the plate surface, which is slightly deeper than a 1/4 t position. Nevertheless, there will still be about 0.5-in. of higher strength and toughness material on one side of the specimen.

Weldment Study of ASIM A533-B

The materials being evaluated are the 12-in.-thick A533 Grade B Class I submerged arc HSST weldment sections

Neg 7017913-1

FIGURE 6.1. HSST Plate 02 A533-B, Class I

designated 51A and 54. The utilization of 1T CT and double cantilever beam (DCB) specimens to determine the toughness profile through Section 51A has been previously described. (1,2) The notches of the CT specimens are parallel to the weldment located in the five following areas:

- a the weld metal center,
- the weld-dilution region on the weld-side of the fusion line,
- a the region of austenitic grain growth on the HAZ-side of the fusion line,
- the center of the HAZ, and
- a the outer boundary of the HAZ.

Nine specimens have been machined from each of these areas and four specimens from each of the A, B, and C areas have been irradiated at 540 °F to 1.5 to 2.5×10^{19} n/cm². Some pre-irradiation fracture toughness properties in areas A, B, and C are listed in Table 6.2 and compared with HSST base plate 02 in Figure 6.2. The DCB toughness values for fracture perpendicular through the weld metal compare favorably with the 1T CT results in area A. It is also readily apparent that the transition temperature of the weldment is lower than that of base plate 02.

Commencing with ETR Cycle 108, 4 additional 1T CT specimens from area A of weldment 51A will begin irradiation at 540 °F toward a fluence of 8×10^{19} n/cm². Beginning Cycle 111, 4 specimens from area D of weldment 51A will be irradiated for a fluence of 1.5 to 2.5×10^{19} n/cm². One 4T CT specimen from area A of weldment 54 will be irradiated with the three base plate specimens in the ATR-4T capsule.

Strain Aging Embrittlement

The intent of this study is to determine the deleterious effects of irradiation and thermal aging upon a vessel which has been subjected to a plastic strain at the roots of cracks,

**TABLE 6.2. Fracture Toughness of A533-B Submerged Arc HSST Weldment Section 51A,
Determined with 1T CT Specimens Parallel to the Weldment**

Specimen Identification	Crack Distance ^(a) From Fusion Line, Mils	Test Temperature		Crack Length a, in.	Load lb	K_Q	$K_{Ic}^{(b)}$
		°F	°C			ksi√in.	ksi√in.
Weld Metal Center, A							
51A 5031	A	-250	-157	1.0412	8,640	62.4	62.4
51A 5032	A	-200	-129	1.0317	10,450	74.5	
51A 5033	A	-225	-143	1.0521	7,420	54.6	54.6
51A 5034	A	-275	-171	1.0363	4,480	32.2	32.2
51A 5035	A	-300	-184	1.0420	4,100	29.7	29.7
Weld Dilution Area, B							
51A 5021	5, 9	-175	-115	1.0574	12,400	92.0	
51A 5023	11, 42	-250	-157	1.0549	4,730	35.0	35.0
51A 5025	4, 15	-225	-143	1.0205	7,890	55.3	55.3
51A 5040	0, 50	-200	-129	1.0624	7,340	54.9	54.9
Area of γ Grain Growth, C							
51A 5022	3, 30	-200	-129	1.0346	9,400	64.7	
51A 5024	0, 18	-250	-157	1.0505	4,600	33.8	33.8

a. The distance in mils from the fusion line to the crack tip in areas B and C.

b. Valid K_{Ic} values per the criteria: $a, B \geq 2.5(K_Q/\sigma_{ys})^2$.

FIGURE 6.2. HSST Sub-Arc Weld 51 A533-B, Class

flaws, or notches. The details of the study and its relation to warm prestressing have been previously described.⁽²⁾ Five 1T CT specimens have been strained for 1 hr and 550 °F with stress intensity levels of 35,000 and 60,000 psi/ $\sqrt{\text{in.}}$. These, along with a series of tensile specimens strained 1% and 10% at 100 °F, have been thermally aged for 1000 hr at 540 °F. Similarly strained tensile specimens have been irradiated at 510 °F to fluence levels from 1.5 to 4.4×10^{19} n/cm².

Fatigue Crack Preparation in CT Specimens

In linear elastic fracture toughness determination, the critical stress intensity (K_{Ic}) is that for fracture initiation from a sharp fatigue crack. Hence, the preparation of a fatigue crack in the fracture specimens is a necessary preliminary to actual toughness determinations. Consideration of the loading conditions during such fatigue crack preparation provides an opportunity to extract some information on fatigue crack behavior. To be sure, these loading conditions were not intended to provide pure propagation data, such as the crack growth rate per cycle (da/dn) versus cyclic stress intensity (ΔK) relationship developed by Paris.⁽⁴⁾ Nevertheless, this information does offer some examples of and insight into the practical situation of crack initiation, stalling, and re-initiation, (incubation) as well as propagation.

The details of the fatigue cracking have been previously described.⁽³⁾ The effective crack growth for initiation and propagation in the base material and weldment approached pure crack propagation rates, with a distinct propensity for hindrance. The fatigue crack propagation in irradiated base material specimens has provided an opportunity for a cursory assessment of the irradiation effect on fatigue. The comparison in Table 6.3 is suggestive of a doubling of the effective propagation rate induced by irradiation.

TABLE 6.3. Irradiation Influence on the Effective Fatigue Crack Extension Behavior in 1T CT Specimens of ASIM A533-B from HSST Plate 02

Fatigue Step	Unirradiated		Irradiated 1.5 to $2.5 \times 10^{19} \text{n/cm}^2$	
	ΔK psi $\sqrt{\text{in.}}$	Crack Rate in./cycle	ΔK psi $\sqrt{\text{in.}}$	Crack Rate in./cycle
1 (from machined notch)	26,100	1.22×10^{-6}	26,060	2.44×10^{-6}
2	22,200	1.73×10^{-6}	21,700	2.71×10^{-6}
3	18,000	1.07×10^{-6}	17,200	1.06×10^{-6}
4	13,300	0.13×10^{-6}	12,700	0.62×10^{-6}

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