**NUCLEAR SAFETY QUARTERLY REPORT
NOVEMBER, DECEMBER, 1968, JANUARY, 1969
FOR
NUCLEAR SAFETY BRANCH
OF
USAEC DIVISION OF REACTOR
DEVELOPMENT AND TECHNOLOGY**

MARCH 1969

**AEC RESEARCH &
DEVELOPMENT REPORT**

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The Staff of Battelle-Northwest

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FOREWORD

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

PREVIOUS REPORTS IN THIS SERIES

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TABLE OF CONTENTS

LIST OF FIGURES	vii
LIST OF TABLES	xi
SUMMARY.	1.1
Engineered Safeguard Studies.	1.1
Pressure Bearing Component Evaluation and Monitoring Studies.	1.4
Fixation of Radioactive Residues	1.5
ENGINEERED SAFEGUARD STUDIES.	2.1
Containment Systems Experiment - G. J. Rogers	2.1
Particle Transport to Surfaces by Condensing Steam - L. C. Schwendiman, J. M. Hales, and T. W. Horst	2.58
PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES.	3.1
Crack Detection in Pressure Piping by Acoustic Emission - C. E. Fitch, Jr., P. H. Hutton, and H. N. Pedersen	3.1
ENVIRONMENTAL STUDIES	4.1
Simulation Modeling of Expected Thermal Generation in Selected River Systems - R. T. Jaske and D. E. Peterson	4.1
FIXATION OF RADIOACTIVE RESIDUES	5.1
SPRAY SOLIDIFICATION	5.1
Laboratory Studies	5.1
Engineering Studies.	5.2
Radioactive Demonstration - W. R. Bond, J. H. Hartley, and A. S. Neuls	5.8
PHOSPHATE GLASS SOLIDIFICATION	5.19
Laboratory Studies	5.19
STUDIES ON AUXILIARIES TO SOLIDIFICATION SYSTEMS	5.19
DEVELOPMENT STUDIES OF SOLIDIFICATION PROCESSES	5.21
Heat Transfer Studies	5.21
INTERMEDIATE AND LOW LEVEL WASTE TREATMENT	5.31
Melter Condensate from Phosphate Glass Process. . . .	5.31
EVALUATION OF SOLIDIFIED WASTE PRODUCTS	5.41

Nonradioactive Laboratory Studies.	5.41
Product Measurements, Testing, and Storage.	5.45
HEAVY SECTION STEEL TECHNOLOGY PROGRAM	6.1
Fracture Studies of Pressure Vessel Steels - C. W. Hunter, J. A. Williams and C. L. Hellerich.	6.1
DISTRIBUTION.	7.1

LIST OF FIGURES

2.1	Elemental Iodine in the Main Vapor Space, Run A-3	2.6
2.2	Elemental Iodine in the Main Vapor Space, Run A-4	2.7
2.3	Elemental Iodine in the Main Vapor Space, Run A-6	2.8
2.4	Elemental Iodine in the Main Vapor Space, Run A-7	2.9
2.5	Elemental Iodine in the Main Vapor Space, Run A-8	2.10
2.6	Comparison of Initial Spray Washout of Elemental Iodine with Drop Absorption Models	2.21
2.7	Visibility in Containment Atmosphere, Run A-2	2.25
2.8	Predicted Dependence of Initial Fraction of Iodine Absorbed by Fog on Fog Density	2.27
2.9	CSE Reactor Simulator Vessel and Primary Restraint System	2.31
2.10	CSE Reactor Simulator Vessel with Core Plate, Run B-19	2.32
2.11	Observed and Predicted Pressure History, Run B-19	2.38
2.12	Fluid Temperature at Nozzle H, Run B-19	2.39
2.13	Fluid State at Nozzle H, Run B-19	2.41
2.14	Fluid Level, Run B-19	2.42
2.15	Sample Strain Output	2.44
2.16	CSE Blowdown B-19 Nodal Layout for WATER HAMMER	2.45
2.17	Observed and Predicted Pressure Below Core Plate, Run B-19	2.47
2.18	Pressure Difference Across Plate, Run B-19	2.49
2.19	Pressure on Core Plate, Run B-19	2.50

TABLE 2.1. *Experimental Conditions - CSE Spray Tests*

	<u>Run A-3</u>	<u>Run A-4</u>	<u>Run A-6</u>	<u>Run A-7</u>	<u>Run A-8</u>	<u>Run A-9</u>
Atmosphere	Air	Air	Steam- Air	Steam- Air	Steam- Air	Steam- Air
Temperature, °F	80	80	250	250	250	250
Pressure, psia	14.5	14.5	44	50	48	44
Nozzle type	(a)	(a)	(a)	(a)	(b)	(c)
Drop MMD, μ ^(d)	1210	1210	1210	1210	770	1220
Drop SMD, μ ^(e)	980	980	980	980	650	980
Geom. Std. Dev., σ	1.53	1.53	1.53	1.53	1.50	1.50
No. of nozzles	3	12	12	12	12	12
Spray rate, gal/min	12.8	48.8	49	49	50.5	145
Total spray volume, gal	510	1950	1960	1960	2020	2300
Spray solution	(f)	(f)	(g)	(h)	(g)	(g)

a. *Spraying Systems Co. 3/4 7G3, full cone*

b. *Spraying Systems Co. 3/8 A20, hollow cone*

c. *Spraying Systems Co. 3/4 A50, hollow cone*

d. *Mass median diameter*

e. *Surface median diameter*

f. *525 ppm boron as H_3BO_3 in NaOH, pH 9.5*

g. *3000 ppm boron as H_3BO_3 in NaOH, pH 9.5*

h. *3000 ppm boron as H_3BO_3 in demineralized water pH 5*

3000 ppm boron as H_3BO_3 was used, and the final pH was 5. This solution is of interest because pH may affect the equilibrium between reactive gases, such as elemental iodine, and the liquid. Parsly⁽³⁾ has shown that under some conditions, the pH could be reduced slightly by reaction with volatilized fission product materials, although an excess of sodium hydroxide is usually provided in candidate spray solutions. Thus, Run A-7 provided the extreme case of zero sodium hydroxide and an acidic solution. Boric acid for nuclear criticality control is used in the solutions stored for emergency core cooling in many nuclear power stations. The same solution is also used for the containment spray systems.

In Run A-8 the prime variable was spray drop size. In all previous tests the drop size was 1210 μ mass median diameter (MMD). In Run A-8 it was 770 μ MMD. These drop sizes are determined by the nozzle manufacturer* by an in-flight photographic technique. Also, a different type of spray nozzle was used. The previous tests used solid cone nozzles, while in Run A-8 hollow cone nozzles were used. Both types were obtained from the same manufacturer* so that consistent drop size information was used.

Run A-9 conditions were similar to those in A-6 except for a higher spray flow rate (0.3 gal/min/ft²). Samples from this test are being analyzed, and results will be reported next quarter.

In all the CSE spray tests to date the sprays were not operated continuously but in intermittent short periods. Table 2.2 lists the time intervals during which sprays were operated. The concentration of six types of fission product

* *Spraying Systems Co., Bellewood, Illinois.*

TABLE 2.2. *Duration of Spray Periods for CSE Experiments (a)*

Time After Start of Iodine Release, min						
	Run A-3	Run A-4	Run A-6	Run A-7	Run A-8	Run A-9
First Spray	Start	40	40.5	30	30	20
	Stop	50	50.5	40	40	33
	Duration	10	10	10	10	3
Second Spray	Start	140	140	80	80	55
	Stop	170	170	110	110	58
	Duration	30	30	30	30	3
Third Spray	Start	1473 ^(b)	1205 ^(c)	1565 ^(c)	1323 ^(c)	200 ^(c)
	Stop	1533	1250	1625	1383	260
	Duration	60	45	60	60	60
Fourth Spray	Start	-	-	-	1443 ^(d)	1350 ^(d)
	Stop	-	-	-	1493	1400
	Duration	0	0	0	50	50
					</	

a. Sprays were with fresh solution at room temperature unless otherwise noted.

b. Fresh demineralized water used in Run A-3 only.

c. Recirculated from vessel sump at sump temperature (~ 200 °F).

d. Fresh solution 3000 ppm B, 1.0 wt% $\text{Na}_2\text{S}_2\text{O}_3$, NaOH, pH 9.4, room temperature.

materials in the gas phase was measured before and after each spray period, and in some cases, during spray operation. The gas concentration within each vessel compartment was found to be homogeneous as determined by Maypack sample clusters located throughout the vapor space.

Typical examples of the concentration-time relation are shown in Figures 2.1 through 2.5. These figures show the gas phase concentration of elemental iodine in the main CSE compartment for Runs A-3, A-4, A-6, A-7 and A-8, respectively.

In the absence of continuing source terms, and for the early period of spraying, theory suggests that the concentration-time relationship may be expressed as

$$\frac{C_G}{(C_G)_0} = \exp [-(\lambda_s + \lambda_n) t] \quad (1)$$

where

C_G = gas concentration at any time

$(C_G)_0$ = gas concentration at $t = 0$

λ_s = removal rate constant due to sprays, min^{-1}

λ_n = removal rate constant due to natural processes,
 min^{-1}

t = time, min.

Thus, the slopes in Figures 2.1 through 2.5 represent the experimental measurements of the total removal rate constants, $\lambda_s + \lambda_n$. It must be emphasized that this is true only when there are no sources of elemental iodine in the gas phase. This was probably the case in all tests before and during the first spray period, but there is evidence that later on in the

FIGURE 2.1. Elemental Iodine in the Main Vapor Space, Run A-3

FIGURE 2.2. Elemental Iodine in the Main Vapor Space, Run A-4

FIGURE 2.3. Elemental Iodine in the Main Vapor Space, Run A-6

FIGURE 2.4. Elemental Iodine in the Main Vapor Space, Run A-7

FIGURE 2.5. Elemental Iodine in the Main Vapor Space, Run A-8

tests, desorption from walls and pools provided a source of iodine, and Equation (1) should no longer be considered valid.

The method used to determine λ_s , the removal rate constant for sprays, was as follows. The concentration at the beginning and end of each spray period was estimated by extrapolating the curves during non-spray periods to the time of beginning and ending of a spray period. The reduction of concentration during the spray period was considered to be due to the combined mechanisms of washout by sprays and natural deposition to structural surfaces. The latter was calculated by averaging λ_n before and after the spray period. Then λ_s was calculated by Equation (2).

$$\lambda_s = \lambda_{\text{measured}} - (\lambda_n)_{\text{avg}} \quad (2)$$

Values of the removal rate constants calculated in this manner are listed in Tables 2.3 through 2.9 for the six transport species studied, plus total iodine. Rate constants are expressed in terms of concentration half lives, the time required to reduce the concentration by a factor of two:

$$t_{1/2} = \frac{\log_e 2}{\lambda} \quad (3)$$

The values listed in columns headed "measured" were taken directly from graphs such as those shown in Figures 2.1 through 2.5. The values listed under "spray only" were calculated as explained above.

One of the objectives of the CSE spray program was to obtain experimental verification of theoretical models which can be used to predict concentrations in large nuclear power containment systems which have containment sprays as a safeguard. The five CSE runs reported here are believed to represent a partial fulfillment of this objective.

TABLE 2.3. Removal of Elemental Iodine in CSE Spray Tests

	Gas Concentration Half Life, min									
	A3		A4		A6		A7		A8	
	Meas- ured ^(a)	Spray Only ^(b)	Meas- ured	Spray Only	Meas- ured	Spray Only	Meas- ured	Spray Only	Meas- ured	Spray Only
Before 1st Spray	62	-	66	-	12	-	16.5	-	17	-
During 1st Spray	5.0	5.5	1.4	1.4	1.9	2.1	2.0	2.2	0.63	0.64
After 1st Spray	46	-	(d)	-	260	-	49	-	150	-
During 2nd Spray	5.5	5.9	8.2	8.2	32	35	21	27	30	33
After 2nd Spray	1700	-	>500	-	540	-	600	-	>500	-
During 3rd Spray	20	20	(c)	-	(c)	-	(c)	-	150	-
During 4th Spray	-	-	-	-	-	-	27	-	32	-

a. Observed half life, obtained from Figures 2.1 through 2.5

b. Corrected for natural processes

c. Concentration increased

d. Indeterminate

2.12

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TABLE 2.4. Removal of Particulate Iodine in CSE Spray Tests

Aerosol Concentration Half Life, min											
A3			A4		A6		A7		A8		
Meas- ured(a)	Spray Only(b)	Meas- ured	Spray Only	Meas- ured	Spray Only	Meas- ured	Spray Only	Meas- ured	Spray Only	Meas- ured	Spray Only
Before 1st Spray	63	-	63	-	40	-	22	-	31	-	-
During 1st Spray	11	12.5	2.5	2.6	2.0	2.2	2.0	2.2	2.4	2.6	2.6
After 1st Spray	115	-	220	-	19	-	20	-	53	-	-
During 2nd Spray	12	12.5	9.4	9.6	6.0	7.1	8	8	(c)	(c)	(c)
After 2nd Spray	370	-	>500	-	(c)	-	(c)	-	(c)	-	-
During 3rd Spray	(c)	-	18	18	(c)	-	(c)	-	(c)	-	-
During 4th Spray	-	-	-	-	-	-	(c)	-	86	-	-

-
- a. Observed half life
b. Corrected for natural processes
c. Indeterminate

TABLE 2.5. Removal of Iodine Form Associated with Charcoal Paper in CSE Spray Tests

		Concentration Half Life, min									
		A3		A4		A6		A7		A8	
		<u>Meas- ured</u> ^(a)	<u>Spray Only</u> ^(b)	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>
Before 1st Spray	(c)	-		(c)	-	65	-	33	-	>1000	-
During 1st Spray	12	13		11	11	4.5	5.0	(c)	-	2.5	2.5
After 1st Spray	400	-		(c)	-	(c)	-	62	-	80	-
During 2nd Spray	60	69		160	160	18	18	14	16	13	14
After 2nd Spray	>500	-		>1000	-	>1000	-	600	-	>1000	-
During 3rd Spray	60	60		>1000	-	(c)	-	(c)	-	(c)	-
During 4th Spray	-	-		-	-	-	-	17.4	-	34	-

a. Observed half life

b. Corrected for natural processes

c. Concentration increased

2.14

BNWL-1009

TABLE 2.6. Removal of Methyl Iodide in CSE Spray Tests

		Gas Concentration Half Life, min									
		A3		A4		A6		A7		A8	
		<u>Meas- ured</u> ^(a)	<u>Spray Only</u> ^(b)	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>
Before 1st Spray	(d)	-		(d)	-	(c)	-	(c)	-	>1000	-
During 1st Spray	(c)	-		(c)	-	(c)	-	(c)	-	5	(c)
After 1st Spray	>1000	-		>500	-	(d)	-	(c)	-	(d)	-
During 2nd Spray	100	100		(c)	-	84	84	100	-	47	47
After 2nd Spray	<1000	-		>500	-	>1000	-	(c)	-	(d)	-
During 3rd Spray	150	-		(c)	-	(d)	-	(c)	-	>500	-
During 4th Spray	-	-		-	-	-	-	130	130	65	65

- a. Observed half life
b. Corrected for natural processes
c. Indeterminate
d. Concentration increased

TABLE 2.7. Removal of Total Iodine in CSE Spray Tests

		Concentration Half Life, min									
		A3		A4		A6		A7		A8	
		<u>Meas- ured^(a)</u>	<u>Spray Only^(b)</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>
Before 1st Spray	70	-		75	-	15	-	18	-	19	-
During 1st Spray	6.0	6.6		2.1	2.1	2.6	2.8	3.5	4.0	1.0	1.0
After 1st Spray	60	-		330	-	41	-	65	-	(c)	-
During 2nd Spray	15	17		65	72	36	64	28	36	26	26
After 2nd Spray	>500	-		>500	-	>1000	-	>500	-	(d)	-
During 3rd Spray	140	140		>500	>500	(d)	-	(d)	-	240	-
During 4th Spray	-	-		-	-	-	-	50	50	52	52

- a. Observed half life
b. Corrected for natural processes
c. Indeterminate
d. Concentration increased

TABLE 2.8. Removal of Cesium in CSE Spray Tests

	Aerosol Concentration Half Life, min									
	A3		A4		A6		A7		A8	
	Meas- ured (a)	Spray Only (b)	Meas- ured	Spray Only	Meas- ured	Spray Only	Meas- ured	Spray Only	Meas- ured	Spray Only
Before 1st Spray	57	-	80	-	34	-	65	-	24	-
During 1st Spray	13	15	3.4	3.5	5.6	6.1	4.0	4.2	2.3	2.4
After 1st Spray	115	-	240	-	>1000	-	95	-	170	-
During 2nd Spray	16	17	9.3	9.5	13	13	10	11	12	13
After 2nd Spray	>500	-	>500	-	460	-	150	-	390	-
During 3rd Spray	25	25	50	-	(c)	-	(d)	-	77	-
During 4th Spray	-	-	-	-	-	-	17	-	22	-

-
- a. Observed half life
b. Corrected for natural processes
c. Indeterminate
d. Concentration increased

TABLE 2.9. Removal of Uranium in CSE Spray Tests

		Aerosol Concentration Half Life, min									
		A3		A4		A6		A7		A8	
	<u>Meas- ured^(a)</u>	<u>Spray Only^(b)</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	<u>Meas- ured</u>	<u>Spray Only</u>	
Before 1st Spray	60	-	96	-	27	-	112	-	30	-	
During 1st Spray	24	35	5.3	5.5	7.8	9.1	5.0	5.4	6.0	6.9	
After 1st Spray	100	-	160	-	(c)	-	>500	-	110	-	
During 2nd Spray	24	27	13	14	12.5	12.8	9.0	9.0	13	14	
After 2nd Spray	>500	-	300	-	300	-	>1000	-	200	-	
During 3rd Spray	45	-	45	-	(d)	-	(d)	-	350	-	
During 4th Spray	-	-	-	-	-	-	(d)	-	(d)	-	

- a. Observed half life
b. Corrected for natural processes
c. Concentration increased
d. Indeterminate

For removal of reactive gases, such as elemental iodine, an expression for λ_s is

$$\lambda_s = \frac{6 F k_c \theta}{V d} \quad (4)$$

where

F = total volumetric spray rate, ft^3/min

k_c = gas phase mass transfer coefficient, ft/min

θ = time that spray drops remain airborne, min

V = volume of gas being sprayed, ft^3

$6/d$ = surface to volume ratio for a sphere, ft^{-1} .

This expression assumes that the gas phase is well mixed at all times and that the mass transfer rate is controlled by diffusion from the bulk gas to the surface of the drop. For precise calculations, the effective value of λ_s should be obtained by summing over the spectrum of drop sizes and velocities. It is convenient to use a simplifying assumption that the entire spray can be treated as if it were of uniform size if the proper mean diameter is chosen. Two mean diameters are selected for the following discussion: the mass median diameter (MMD) and the surface mean diameter (SMD).

The time a spray drop remains airborne, θ , can be estimated by dividing the average vertical distance from the spray nozzles to intercepting surfaces, h_{avg} , by the average vertical drop velocity. Drops are created with an initial velocity higher than their average fall velocity and slow down during fall because of viscous drag. The average velocity was estimated by two methods. First, the assumption was made that the drops fall at terminal velocity during the entire fall period. Second, the effect of the high initial velocity was estimated by allowing for drag and assuming the initial velocity equaled the nozzle flow rate divided by the nozzle orifice

cross-sectional area. Spray removal coefficients were calculated by modifying Equation (4) with these assumptions. Equation (5) is the simplest expression, using the mass median diameter and the terminal settling velocity for this diameter, U_t :

$$\lambda_s = \frac{6 F k_c h_{avg}}{V d_{MMD} U_t} \quad (5)$$

In Equation (6), the surface mean diameter is used and the fall velocity is corrected for initial velocity.

$$\lambda_s = \frac{6 F k_c h_{avg}}{V d_{SMD} U_{avg}} \quad (6)$$

A comparison of the spray removal rate constants predicted by Equations (5) and (6) with the experimental results from Table 2.3 elemental iodine is shown in Figure 2.6. The experimental results are those obtained from the first spray period. The most important conclusion is that agreement between measured washout rates and predictions based on simple washout models is within $\pm 20\%$. As would be expected, using the surface mean diameter predicts faster removal than that predicted using the mass median diameter. The difference is not great, however, and the use of either one of these mean diameters is probably acceptable in view of uncertainties in drop size data and other parameters incorporated in the models.

Several other conclusions can be drawn from Figure 2.6. Reference to Table 2.1 shows that the only difference between Runs A-3 and A-4 was that Run A-4 had 3.83 times greater spray flow rate. From Figure 2.6, λ_s is 3.93 times greater for Run A-4 than A-3: a good agreement. Run A-9 will provide a further check on flow rate at still higher flows.

FIGURE 2.6. Comparison of Initial Spray Washout of Elemental Iodine with Drop Absorption Models

Another conclusion is that the model accounts adequately for differences in atmospheric properties. Runs A-4 and A-6 were duplicates except that room temperature air was used in Run A-4 while a saturated steam-air mixture at 250 °F and 44 psia was used in Run A-6. The large difference in atmospheric conditions caused only a slight difference in the spray removal coefficient, as predicted.

The effect of drop size can be seen by examining Runs A-6 and A-8. Good agreement with theory is evident.

The last conclusion is that pH of the spray solution had only a minor influence on the spray removal rate constant. Run A-6 used a solution with excess sodium hydroxide and a pH of 9.5. Run A-7 had no caustic, and the pH was 5. The fact that the removal rate was essentially independent of pH suggests that reactions other than simple hydrolysis⁽⁴⁾ occurred. The excellent agreement between theory as predicted by Equations (5) and (6), and experiment shows that elemental iodine was transferred to the aqueous drops with negligible liquid phase resistance. Thus that any chemical reactions must be very rapid. Further study of chemical reactions between iodine and aqueous sprays is warranted to provide better understanding of the observed behavior.

Equations (5) and (6) have been shown to be valid expressions for the rate constant for the range of λ_s up to about 1 min^{-1} . This is an important conclusion, but it is not sufficient in itself to enable predictions of the complete time dependence of elemental iodine concentration in the gas phase. The proper rate equation must be provided. Equation (1) is the solution of a simplified rate equation applicable in the absence of source terms and for the case of negligible liquid phase concentration. It should not be used for times

greater than that required to reduce the gas phase concentration of elemental iodine to about 1% of its initial value. At longer times iodine in the liquid pool or in the liquid film on structural surfaces will diffuse back into the gas phase. This may be true even if fresh sprays with zero concentration of iodine are used. This quasi-equilibrium can be expressed by the approximate equation

$$\frac{C_G}{(C_G)_0} \approx \frac{V}{LoH + Fht} \quad (7)$$

where

Lo = liquid volume in system at time sprays started

$$H = \frac{C_L \text{ total iodine}}{C_G \text{ elemental iodine}} = \text{the equilibrium gas-liquid distribution coefficient.} \quad (8)$$

Evidence of this equilibrium behavior can be seen in all of the CSE spray tests. The gas phase concentration was not decreased in second and third spray periods to the extent that it was in the first period.

Effect of Fog Formation on Transport of Iodine -

J. G. Knudsen* and R. K. Hilliard

Nine large-scale fission product transport experiments have been made in the main CSE vessel to date. With the exception of the first experiment, visual observations were made throughout the entire 20 to 30 hr test durations. Three atmospheric conditions were used: (1) air at 75 °F, 85% relative humidity, 14.5 psia, (2) saturated air-steam mixture at 180 °F, 23 psia, and (3) saturated air-steam mixture at 250 °F, 48 psia. A similar pattern of events was observed

* Oregon State University, Corvallis, Oregon

regardless of the atmospheric conditions employed. Prior to release of iodine or aerosol materials, the atmosphere was clear and visibility was good. Releasing fumes from oxidizing, molten Zircaloy-clad UO_2 had no appreciable effect on visibility. However, as soon as iodine and cesium release started, fog formed near the point of release and spread rapidly throughout the containment vessel. The visibility rapidly decreased to about 1 ft. If sprays were not operated, the visibility remained at this low value for about 30 min, then gradually improved, returning to its initial condition in 3 to 5 hr. Figure 2.7 gives the estimated visibility as a function of time for Run A-2. Operation of sprays for as little as 3 min cleared the atmosphere of fog.

The fog is believed to form by condensation of water vapor on nuclei of cesium oxide particles. Iodine vapor alone has been observed to cause fog in small-scale tests in the Aerosol Development Laboratory (ADF). Whether the nucleating matter was iodine, cesium, or both, it is very probable that iodine is rapidly absorbed in the fog droplets to its equilibrium concentration. This has two effects on iodine transport. First, the concentration of elemental iodine is reduced almost instantaneously by an amount commensurate to the mass absorbed, and the concentration of particulate-associated iodine is increased by the same amount. Second, the time dependence of both forms may be altered by desorption from the fog droplets at later times.

The first effect has been noted many times in small-scale tests in the ADF, as well as in the large-scale CSE tests. When small masses of iodine are released, resulting in initial gas phase concentrations of about 1 mg/m^3 , a large fraction is found on the Maypack filters. Even after correcting for the known absorption of elemental iodine on filter media, this

FIGURE 2.7. Visibility in Containment Atmosphere, Run A-2

fraction often exceeds 50% of the combined iodine forms. However, when a larger mass of iodine is released, giving about 100 mg/m^3 initially in the gas phase, only a small fraction is found to be associated with the filters. These observations are consistent with an equilibrium situation between I_2 in the gas phase and absorbed iodine in all forms if the equilibrium gas-liquid distribution coefficient is concentration dependent. Eggleton⁽⁴⁾ has published theoretical curves of gas-liquid distribution coefficient versus liquid concentration which suggest that this is the case. For concentrations in the range of interest, his data can be expressed as

$$C_G = K C_L^2$$

where

C_G = gas phase concentration of elemental iodine

C_L = liquid phase concentration of all iodine species

K = a constant.

The fraction of the iodine released to containment which is initially absorbed by fog droplets is proportional to the liquid water content of the fog and inversely proportional to the iodine concentration. The predicted relationship is shown in Figure 2.8 with water content of the fog being the independent variable and iodine concentration the parameter. The liquid content of the fog observed in the CSE tests was not determined, but the very low visibility shown by Figure 2.7 is compatible with liquid contents in the range of 100 g/m^3 .⁽⁵⁾ A liquid content of about 500 g/m^3 would be required to explain the Maypack determinations, according to Figure 2.8.

FIGURE 2.8. Predicted Dependence of Initial Fraction of Iodine Absorbed by Fog on Fog Density

Particle Size Analysis for CSE Spray Tests - L. F. Coleman

Particle size measurements have been obtained for CSE spray Runs A-4, A-6, A-7, and A-8. Measurements for Run A-4 were obtained by sampling outside the containment vessel with a Cassella* impactor. Subsequent sampling during pressurized steam-air tests (Runs A-6, A-7, and A-8) was performed by insertion of a Scientific Advances** inertial impactor directly into the 250 °F, 48 psia containment vessel atmosphere. The inertial impactor was heated to 300 °F prior to positioning in a pressure chamber which communicates with the containment vessel via a ball valve. After 1 min thermalization, the impactor is inserted into containment and sampling is commenced. Sampling rate was controlled by a limiting orifice following the sixth and final stage of the impactor. Stage cut off limits for unit density particles are 0.25, 0.5, 1.0, 2.0, 4.0, and 8 μ and greater for the six stage impactor.

Size data from the four tests where measurements were taken are given in Table 2.10.

The particle size distribution for cesium deviated from a log normal type distribution in most cases, being skewed at the small particle size end of the curve. One interpretation of this could be that the small cesium particles grow rapidly by steam condensation and also are removed more efficiently by diffusion during spray periods.

In general, the aerodynamic mass median diameter for both cesium and uranium is reduced in magnitude following the spray periods. Also, there is no gross difference in particle size

* C. F. Cassella & Company Ltd, London, England

** Scientific Advances, Inc., Columbus, Ohio

distribution and particle content prior to and after recirculation of spray solutions. Recirculating spray systems might be expected to produce some aerosol droplets. However, this should be a self-limiting process with only an extremely small percentage of the released aerosol mass being left in the gas phase after spraying ceases.

Particles of the general size given in Table 2.10, when used in spray washout models, give lower removal rates than that actually observed in CSE tests. J. D. McCormack⁽²⁾ has previously discussed the effects of particle size on washout by sprays.

TABLE 2.10. *Particle Size Analyses - CSE Spray Tests*

	Aerodynamic Particle Diameter ^(a) , μ MMD							
	Run A-4 ^(b)		Run A-6 ^(c)		Run A-7 ^(c)		Run A-8 ^(c)	
	Cs	U	Cs	U	Cs	U	Cs	U
Before 1st Spray	0.9	1.6	0.5	1.0	2.0	1.8	0.5	0.8
After 1st Spray	0.5	0.7	0.4	0.9	0.3	0.6	0.6	0.9

-
- a. Diameter relative to settling velocity of a unit density spherical particle.
- b. Cassella Impactor; sampled outside the containment vessel.
- c. Scientific Advances Inertial Impactor; sampled within containment vessel.

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

A reactor simulator vessel with a volume of 150 ft³ and 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 nozzles directed horizontally as shown in Figure 2.9. 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 vessel, thrust reaction forces, shell stresses, and forces on reactor dummy core structures.

Installation of instrumentation in the test apparatus was completed and a blowdown test series was begun. The purpose of the current series is to study the effects on the subcooled and saturated phases of coolant blowdown of a simple perforated plate separator representative of mixer, support, and grid plates used in nuclear reactors. The first test (B-19) of the series was conducted with a small break (1.687 in. break diameter) and the sieve plate from subcooled initial conditions (2150 psig, 600 °F).

Experimental Arrangements - A. J. McElfresh and
M. E. Witherspoon

Facility and Vessel Configuration. Blowdown (B-19) was made from the bottom nozzle of the CSE simulator vessel with an elbow, extension, and orifice plate as shown in Figure 2.10. The core plate divided the vessel volume to give a ratio of upper to lower volume of 2.3. Sixty four nozzles of 1.23 in.²

Neg 0690250

FIGURE 2.9. CSE Reactor Simulator Vessel and Primary Restraint System

each were installed with the remaining 24 holes plugged. The flow area of this sieve plate was 0.55 ft^2 . The break area in the discharge orifice of 0.0156 ft^2 results in a break area to free plate area ratio equal to 0.0282. Blowdown initiation was accomplished using the double-rupture disk, over-pressurization technique.

Figure 2.9 shows the general layout of the primary and secondary restraint systems. The primary system is used for measurement. The secondary system provides backup support should a gross failure occur in the primary system. Cables are used in the secondary system to provide vertical and horizontal restraint. Sufficient slack in the vertical restraint cables was not provided prior to vessel heatup for Run B-19. Resultant vessel expansion during heatup exceeded the free limits of two of the vertical restraint cables. This resulted in a preload on the primary system load cells so no useful thrust or weight data were obtained from this test.

To make sure that the vessel was completely full of water, vent lines were connected to all simulator top nozzles in which gases could be trapped. The vent lines were joined and then passed through a heat exchanger and to a (2.6 gal) accumulator. The other end of the accumulator was attached to the nitrogen pressurizing system. Thermocouples were used to determine liquid presence in the accumulator.

Experimental Instrumentation. Instrumentation for the blowdown runs is divided into two primary sensor groupings. The first group consists of those sensors necessary for the measurement of blowdown force and fluid dynamics. The second group consists of instrumentation installed for the

purpose of monitoring potential propagation of a defect indication zone in the lower section of the pressure vessel.⁽⁶⁾

Table 2.11 is a summary of the two groupings. All of these sensing devices are connected to the input of a small digital computer for data logging. The outputs of all sensors are read from 5 to 50 times per second during the blowdown period. Sixteen of the sensors listed in Table 2.11 were selected for high speed scanning on a second computer device. All the pressure sensors, two of the core plate bolts and the weight-of-vessel-contents load cell were scanned at a rate greater than 3000 points/sec. The nine acoustic emission sensors were recorded separately on an analog tape recorder. Output from all sensors scanned by the small digital computer is reduced to plots via routines developed for a UNIVAC 1108 system. These plots are designated as "HI-SPEED" when the scan rate is 3000 points/sec.

Pressure and temperature sensors are located in nozzles J, H, R, (refer to Figure 2.10) the instrument ring, and in taps on the nozzle extension piece. Additional thermocouples are located along the length of the liquid level probe and are designated TL1 and TL3 to TL6 in the figure. A special thermocouple assembly is installed in Nozzle "O" for transient heat transfer measurements. Four of the eight support lugs for the dummy core plate are instrumented with strain bolt assemblies for measuring the integrated force on the plate. The center of the core plate and one of the lugs also have strain gages installed. Additional strain gages are installed on the inside and outside of the area about the base of Nozzle C. Each of these strain gages has a thermocouple attached to the vessel skin in its vicinity. A differential pressure sensor is attached to tubing routed through the instrument ring to one of the orifices inside the vessel. A second

TABLE 2.11. *Blowdown Run B-19 Instrumentation*

Group I - Sensors for Blowdown Variable

<u>Number</u>	<u>Description</u>	<u>Purpose</u>
1	Strain gage load cell	Weight of vessel contents during blowdown
2	Strain gage load cells	Thrust forces during blowdown
1	Liquid level probe	Height of water in blowdown vessel
7	Strain gage pressure sensors	Pressure at various vessel nozzles
3	Strain gage pressure sensors	Pressure inside vessel, below core plate
2	Strain gage differential pressure sensors	Flow rate through core plate, and through nozzle extension
16	Calibrated thermocouples	Temperature of vessel fluid
1	Strain gage on center of core plate	Force on core plate
8	Strain gage on core support bolts	Force on core plate
1	Strain gage on core support lugs	Force on core plate
1	Linear displacement potentiometer	Nozzle deflection during blowdown

Group II - Sensors for Monitoring the Defect Indication

13	Thermocouples	Temperature of fault zone metal inside
15	Thermocouples	Temperature of fault zone metal outside
26	Strain gages, outside metal surface	Stress map of fault zone
3	Strain gages, inside metal surface	Stress map of fault zone
9	Acoustic emission sensors	Acoustic emission of fault zone

differential pressure sensor is attached to pressure taps 10 in. apart in the center of the nozzle extension piece. A linear displacement device is installed on the bottom of the nozzle extension piece to monitor the action of the nozzle during blowdown. In addition to these sensors, acoustic emission transducers are installed on the top and bottom vessel head for the purpose of monitoring any defect propagation in the bottom head.

Run Conditions for B-19. The initial simulator pressure was 2150 psig, about 572 psi above the saturation pressure at the initial nominal fluid temperature of 600 °F. The loop was pressurized to prevent boiling in the heat exchanger during heatup, while water was circulated up through the simulator vessel. The beginning vessel temperature gradient from top to bottom was about 70 °F at a nominal 250 °F average vessel temperature. As the heatup progressed, the gradient was 15 °F at 400 °F, 12 °F at 525 °F and less than 1 °F at 592 °F. The circulation scheme used and natural convection are credited for the quite homogeneous fluid mixture attained.

A "dead leg" of cooler water in the blowdown nozzle extension existed in earlier experiments. Modifications to the nozzle extension were made to permit bleeding cooler water from this dead leg, thereby raising the temperature of the water in the nozzle extension. When the nozzle extension temperature was no longer changing, the vessel was isolated from the heating loop. The nitrogen pressure in the accumulator was reduced to 2150 psig by venting. The data logging system and rupture disc mechanisms were then triggered to start the blowdown.

Preliminary Results - R. T. Allemann

Saturated Blowdown Analysis. Figure 2.11 shows the pressure inside the vessel versus time predicted by the FLASH-2 code. For code input, the vessel was divided into two lumped volumes. Pressure in the volumes above and below the sieve plate was calculated as a function of time. The figure also shows the experimental data from the pressure sensor located in Nozzle H above the core plate.

The pressure data illustrates an initial transient decompression, then a general decompression out to 52 sec, and a change in the decompression slope when the fluid reaches the blowdown nozzle and the blowdown enters the final high quality discharge stage. Similarly, the plot of temperature in Figure 2.12 at the same location follows the decompression of the fluid to the high quality transition, changes slope during the final blowdown stage, equilibrates at 212 °F while the vessel is boiling off the remaining water in the bottom of the vessel, and begins rising back to the metal temperature as metal heat is given up to the superheated steam remaining in the vessel. It is obvious from the pressure plots that the coefficient applied to the FLASH-2 Moody leak rate calculation gives the correct "knee" in the pressure curve. The coefficients used were the same as those determined from previous bottom blowdowns through a 1.689 in. orifice. It is concluded that the sieve plate did not greatly change the average blowdown rate for the small bottom break from that of a vessel without internals.

The outstanding feature of the curves in Figure 2.11 is the duration of the subcooled regime. The FLASH-2 prediction was based on the assumption of saturated fluid in the vessel and a 500 msec decompression time.

FIGURE 2.11. Observed and Predicted Pressure History, Run B-19

FIGURE 2.12. Fluid Temperature at Nozzle H, Run B-19

Figure 2.13 shows that in spite of an effective temperature-pressure error of 30 °F, the change in slope indicates that saturation did not occur in Nozzle H until 25 sec of blowdown had elapsed. Because special precautions had been taken to ensure that the vessel was completely full of water, it is difficult to explain the long subcooled period. An earlier run made without internals in the vessel (B-17, 1.689 in. D bottom nozzle, 2375 psig, 600 °F) showed a saturation time of 26 to 30 sec. The long subcooled time of B-17 was attributed to a possible steam dome in the top of the vessel resulting from incomplete filling. The vessel for Run B-19 was believed to be completely full. This was insured by having liquid in the pressurizer tank above the vessel. Another run will be made to check the initial conditions and eliminate any possibility of operations error in establishing the initial condition.

Other possible causes of the long duration subcooled blowdown phase might be local boiling at the hot wall of the vessel or nitrogen solubility. Calculations are being made to check these hypotheses. If further blowdown experiments bear out the long duration "subcooled" regime, it could have very significant effects on the safety calculations for real reactors. The long "subcooled" period may mean that the pressure oscillations continue for some time (i.e., many cycles) and resonance effects would be of importance. Other results here indicate that the long decompression duration is accompanied by smaller amplitude pressure fluctuations than those predicted.

Figure 2.14 shows the output from the TDR (Time Domain Reflectometer) level device. Two liquid level plots are shown, one of which is from an earlier 2 in. blowdown (Run B-17) without a dummy core plate in the vessel. The vertical liquid level drop which is seen on the B-19 signal and is not seen

FIGURE 2.13. *Fluid State at Nozzle H, Run B-19*

FIGURE 2.14. Fluid Level, Run B-19

on the B-17 plot occurs at the level of the core plate installed for B-19 and suggests that a high quality gas dome exists just under the core plate. Both blowdowns enter the high quality transition stage of blowdown at nearly the same time.

The data in Figure 2.15 show that the strain gage on a core support bolt inside the vessel has relatively small changes in the level of indicated strain until the transition period, when it is influenced by the rapid change in temperature of the fluid. As the remaining steam is superheated and temperature of the vessel vapor contents and the bolt reach equilibrium, the indicated strain approaches its original value.

Subcooled Blowdown Analysis. The decompression of the fluid in the vessel was calculated using WATER HAMMER.⁽⁷⁾ Earlier tests with this code had shown that high frequency oscillations (750 to 1000 Hz) accompanying the decompression could be generated by inclusion of the various 8 and 4 in. nominal diameter nozzles on the vessel. Some of these nozzles have blank flanges and some have a pressure gage or thermocouple mounted on the flange. The 2 in. nozzles and pipes were not included in the WATER HAMMER solution. Figure 2.16 shows the nodal arrangement and temperature profile used to represent the B-19 blowdown. Thirty eight nodes were used. Table 2.12 gives the input values for these nodes. The sieve plate region was represented by only one node because earlier calculations had shown that more nodes did not improve or change the pressure drop prediction.

Figure 2.17 shows the pressure predicted by the subcooled WATER HAMMER code versus time compared to some measured pressures. The outstanding differences are that the actual

FIGURE 2.15. Sample Strain Output

FIGURE 2.16. CSE Blowdown B-19 Nodal Layout for WATER HAMMER

TABLE 2.12. Nodal Input Values - WATER HAMMER

No.	Blowdown Run B-19					Nozzle Designation
	A	X	a	Temp	τ	
1	0.253	0.261	3635	502	7.2	
2	↓	1.01	↓	502	28	
3	↓	↓	↓	502	28	
4	↓	↓	↓	502	28	
5	↓	↓	3445	525	29	
6	↓	↓	3000	575	34	
7	6.50	1.00	2750	601	36	
8	9.35	↓	2750	601	36	
9	↓	↓	2740	602	37	
10	↓	↓	2740	602	37	
11	↓	0.9723	2745	603	26	
12	↓	0.0273	2730	604	1	
13	0.559	0.338	2770	600	12	
14	9.35	0.0273	↓	↓	1	
15	↓	0.8727	↓	↓	31	
16	↓	1.00	↓	↓	36	
17	↓	↓	↓	↓	↓	
18	↓	↓	↓	↓	↓	
19	↓	↓	↓	↓	↓	
20	↓	↓	↓	↓	↓	
21	↓	↓	↓	↓	↓	
22	↓	↓	↓	↓	↓	
23	↓	↓	↓	↓	↓	
24	↓	↓	2750	602	36	
25	6.3	↓	2740	602	37	
26	3.0	0.33	2740	602	12	
28	0.253	1.35	3000	575	45	D
29	0.0643	1.15	2750	601	42	J
30	0.253	1.43	2750	601	42	B
31	0.0643	1.15	2750	601	52	S
32	↓	1.43	2760	601	52	H
33	↓	1.15	2775	599	41	G
34	↓	↓	2775	599	41	O
35	↓	↓	2750	601	42	Q
36	↓	↓	2750	601	42	R
37	0.253	1.43	2750	601	50	A
38	0.253	1.43	2750	601	50	E

A = Nodal area, ft^2

X = Nodal length, ft

a = Local sound velocity, ft/sec

T = Local temperature, $^{\circ}\text{F}$ τ = Nodal time of wave transverse, 10^{-5} sec

FIGURE 2.17. Observed and Predicted Pressure Below Core Plate, Run B-19

oscillations and the major pressure steps are also smaller than predicted. The latter result implies a longer decompression time than calculated. The lower amplitude, about 1/3 of the calculated value, implies that the pressure drop across the sieve plate or other structure is less than calculated by WATER HAMMER.

The pressure drop across the plate predicted by WATER HAMMER versus time is shown in Figure 2.18. Also shown are pressure drop predictions made for a larger break and plate area but for the same temperature. The predictions have been scaled down on the basis of the area ratios involved to apply broadly to the 1.689 in. D blowdown. The amplitudes obtained by this method cannot be expected to give agreement since not all area ratios were invariant. However, agreement in frequency should have been obtained. The run data for comparison with these predictions are very sparse because of the low values involved and a pressure sensor malfunction. The difference between two sensor readings such as in Figure 2.17 has large uncertainty because the error band is ± 30 psi.

Figure 2.19 is a plot of output from one of the bolts which measures force on the core plate. The output of this bolt was calibrated before the run by applying known loads to the plate and relating the force to the equivalent pressure on the surface of the plate. The magnitude of the pressure on the plate as determined from the calibration is shown. An additional coefficient of 1.5 times this reading should be applied to account for the difference between central loading during the calibration and the uniform loading experienced during the run.

The data show plateaus resulting from the digitization of minimum level output voltages. Included on this same plot is

FIGURE 2.18. Pressure Difference Across Plate, Run B-19

FIGURE 2.19. Pressure on Core Plate, Run B-19

the expected pressure on the plate from an output of the code WATER HAMMER. Note that the scale for the code output is 5 times that of the measured pressure. An appropriate amplitude reduction factor may have to be applied to WATER HAMMER in order to resolve the prediction error (barring the hypotheses discussed above and/or unknown calculational error). Frequency agreement between the measured and actual pressure drop prediction is fair, but the complexity of the wave shape will be extremely difficult to model accurately and such inaccuracies will have to be allowed for by conservative design.

Conclusions. Results from Run B-19 showed a slower rate of decompression and less severe pressure oscillations than predicted in the plate region. However, there were some local high differential pressures noted. The subcooled conditions lasted longer than expected. The sieve plate allowed a second steam dome to form before it during the saturated regime of blowdown, but did not have a significant effect on the overall blowdown time when compared with data from a vessel without the dummy core plate.

Simulator Vessel Defect Surveillance - W. C. Townsend

Ultrasonic inspection of the CSE reactor simulator vessel following a structural failure of the support system revealed a defect indication in the vicinity of the bottom blowdown nozzle. The defect indication exceeded the size allowed by Section III of the ASME Pressure Vessel and Boiler Code. Core samples were removed from the defect region, and extensive tests were made on these samples. Test results showed that the defect indication was the result of clouds of small silicon and aluminum oxide particles, and that the base metal was of normal strength and ductility. Results from the core sample tests and from detailed stress analyses indicated that the

simulator vessel was still serviceable for blowdown experiments. Accordingly, the blowdown program was resumed without repair of the defect indication, but extensive monitoring of the defect region is being performed to detect any change in size or characteristics of the defect indication, and to obtain stress and temperature data to confirm the analytical studies. The ultrasonic inspections are in compliance with ASTM A-388 and ASTM A-435, and Section III of the ASME Pressure Vessel and Boiler Code. The defect region is extensively instrumented with strain gages and thermocouples to monitor temperatures and strain of the defect region. This instrumentation and the necessary cables prevented surveying the entire defect region ultrasonically.

Inspections were performed following loading tests made on the nozzle by placing axial and bending moment loads on the nozzle and following a hydrostatic pressure test. There was no indication of defect growth from any of these tests. After blowdown experiment B-19 (2150 psig, 600 °F) was conducted, the defect region was ultrasonically inspected per the above procedures.

The areas inspected are shown shaded in Figure 2.20. No change of the defect indication was found. Acoustic emission monitoring was also performed during the nozzle loading tests and blowdown B-19. No significant emissions were detected.

Temperature data were obtained from internal and surface thermocouples. Location of these thermocouples is shown in Figure 2.21. Temperature history plots for TIFZ3, 4, 5, and 6 are shown in Figure 2.22. These data indicate a maximum temperature gradient of approximately 270 °F in 1.075 in. in the thermocouple plug. This maximum gradient occurs at 128 sec after the blowdown starts. At this time the water in

FIGURE 2.20. *Fault Zone Region in Lower Hemispherical Head of CSE Simulator Vessel*

FIGURE 2.21. Thermocouple Location in Fault Zone Region

FIGURE 2.22. Temperature Profiles in Fault Zone, Run B-19

the bottom spherical head of the vessel is below the bottom of the blowdown nozzle and the vessel pressure is atmospheric. Therefore, the water temperature is at 212 °F. The measured temperature gradient is greater than that predicted, based on an inside film coefficient of 700 Btu/hr-ft²-°F. Detailed thermal analyses based on the experimental temperature data and analyses of strain data are continuing.

Data Acquisition and Processing - N. P. Wilburn

Processing of Aerosol Data

A code "AERDP" has been written for the Univac 1108 computer to process data collected using a PDP-7 as a multi-channel analyzer for an aerosol transport run of the "A" series.⁽⁸⁾ Samples containing ¹³¹I, ¹³⁷Cs, etc. from Maypack clusters, deposition panels, liquid collection troughs and sumps are analyzed. Data from the samples is written onto a "DECTAPE" magnetic tape intermediate storage media as it is collected.⁽⁹⁾ It is then transferred to a standard 1/2 in. wide IBM compatible tape using a recently revised code.⁽¹⁰⁾

AERDP uses the IBM tape as input, processes it, and then prints out information about the Maypack, deposition, and liquid samples. Averages over the samples collected at a given time, associated standard deviations, and miscellaneous derived quantities are computed. Corrections can be applied to the data as it is processed. The printed report tabulates concentrations of the specified isotopes, analytical errors, percent of the total specified isotope present in the sample, standard deviation of all samples from average and deviation of sample value from average expressed as number of standard deviations. All of these are given as functions of time at which the sample was collected.

In addition to a printed report, AERDP also plots the processed data under control of the input deck. For instance, in the case of the Maypack data, plots are prepared which are very similar to those given in this quarterly report, in that logarithm of the isotopic concentration is plotted as a function of time. In the future some of these plots will probably appear herein.

The average processing time on the 1108 computer for 1000 samples is 1.2 min. During a typical aerosol run, 1500 to 2000 samples of various types are collected and analyzed.

Processing of Blowdown Data

A UNIVAC 1108 "MPXDP" for processing the data collected during blowdown experiments has been described in a previous quarterly report.⁽¹¹⁾ Printed reports of the data collected by the data logging system are given as a function of time by the code "MPXDP." In addition, plots are prepared for the 3 to 10 min duration of the run.

During this quarter, some additional features have been added to the code. First, plots of the data collected by the high speed multiplexer for the subcooled portion of the blowdown are now prepared under control of the input deck. Second, the processed data can be written out to drum or tape for additional processing by other codes such as "DENPRO" which was described last quarter.⁽¹²⁾ Third, plots of the derivatives of the data can now be made.

A new code, "CRSPLT", has been written to use the second feature above to prepare cross plots of the blowdown data. For example, this code will be used to compare the pressure versus temperature history at a point in the vessel during a run with the known saturated curve.

Blowdown Data Logging System

For the experimental runs up through B-18, "time zero" (the time the inner blowdown disc ruptured) was assumed to be when the wires attached to the outer of the two rupture discs broke. From analysis of the data collected during these runs, it was determined that the wires were breaking 50 to 100 msec or more before the initial pressure drop occurred just inside the inner disc. Since logging by the high speed multiplexer was initiated at "time zero", and occurs only for 100 msec, much or all of the subcooled data was usually lost.

Circuitry was added to the system for Run B-19 to allow "time zero" to be that when the initial pressure drop occurred. The subcooled portion of B-19 was adequately logged.

However, in order to obtain information as to the length of time it takes for the rupture of the disc to occur, break wires will be put on the inner disc and then will be used to trigger a memory scope which will monitor the same pressure drop which determines "time zero." This system will be operational for Run B-20.

PARTICLE TRANSPORT TO SURFACES BY CONDENSING STEAM -

L. C. Schwendiman, J. M. Hales, and T. W. Horst

Introduction

The objective of this investigation is the development of a quantitative understanding of the processes causing particle capture in condensing-steam atmospheres, with special reference to post-accident reactor containment systems. The study was planned to approach the problem on a theoretical and on an experimental basis; progress in each of these areas is reported.

Theoretical Modeling

The analysis of the laminar, condensing-steam boundary layer was extended to allow for variations in viscosity and specific heat. The equation describing aerosol transport in such a system was revised to the form,

$$\frac{\beta Sc}{v_{\infty}} \gamma'' = \Sigma_1 \gamma' + \Sigma_2 \left(\gamma + \frac{N_i}{N_{\infty} - N_i} \right) ,$$

where

$$\Sigma_1 = -\phi_{\mu}^2 \pi [3ScF + (\Delta W) q \xi'] + \frac{2\beta Sc \phi_{\mu}'}{v_{\infty} \phi_{\mu}} ,$$

and

$$\begin{aligned} \Sigma_2 = -\phi_{\mu}^2 \left(\pi' [3ScF + (\Delta W) q \xi'] \right. \\ \left. - \pi \left\{ [(\Delta W) q \xi']^2 - (\Delta W) q \xi'' \right\} \right) . \end{aligned}$$

N_i = interfacial particle population density

N_{∞} = bulk particle population density

$$\gamma = \frac{N - N_i}{N_{\infty} - N_i}$$

$$q = \left(W + \frac{1}{\sqrt{M_A/M_W} - 1} \right)^{-1}$$

ρ = gas density

ρ_{∞} = gas density in bulk

M_A = molecular weight of air

M_W = molecular weight of water

W = mass fraction, water

F = dimensionless variable defined by Minkowycz and Sparrow⁽¹³⁾

$$\xi = \frac{W - W_i}{W_\infty - W_i}$$

Sc = Schmidt number, ν/D_{AB}

$\beta = \phi_\mu D$, where D = particle diffusivity

$$\phi_\mu = \mu/\mu_\infty$$

$$\pi = \frac{\rho_\infty}{\rho}$$

μ = gas viscosity

μ_∞ = gas viscosity in bulk.

The revised system of equations was programmed on the hybrid computer. At present the program is nearly ready for solution of the equation, final interfacing being required for its completion.

Upon completion of the laminar boundary-layer analysis, the present investigation will be extended to consider the turbulent regime.

Experimental Work

The experimental phase of this investigation has been limited to the preliminary design of the vessel required, some detailing of the essential features, and acquisition of sensitive anemometer and temperature probes.

Work has been limited to this extent necessarily due to reduction in the funds originally requested for this study. Since the work was initiated and has been sponsored by the joint AECL-USAEC Heavy Water Reactor Cooperative Program, consideration has been given to having the experimental work continued by scientists at Chalk River. This was suggested

as a possible alternative because of the phasing out of the Cooperative Program and the possibility that facilities may be readily available at Chalk River for carrying out the experiments.

With the anticipation that the AECL may wish to pursue the experimental approach, we submitted drawings of the apparatus and a description of the research plan.⁽¹⁴⁾

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3. PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

CRACK DETECTION IN PRESSURE PIPING BY ACOUSTIC EMISSION - C. E. Fitch, Jr., P. H. Hutton, and H. N. Pedersen

Introduction

In the interest of further enhancing nuclear reactor safety, the USAEC Division of Reactor Development and Technology has been sponsoring a program at The Pacific Northwest Laboratory since February 1966 to develop application of "acoustic emission" to monitor for flaw growth in nuclear reactor pressure boundaries.

"Acoustic emission" represents a new and unique technique for detecting flaw growth in solids by sensing the elastic waves generated in the material from energy released by the fracture process.

Related work is also being sponsored by DRDT with Phillips Petroleum Co., Idaho Test Site on plant application of developed techniques.

See References 1 to 5 for reports and papers published.

Discussion

Program developments are discussed under the topical outline as used in previous reports.

Significantly reduced FY-69 funding for this program has required serious curtailment of work. This is reflected in

some of the objectives for the quarter such as environmental testing of transducer materials not being accomplished.

Task 1 - Acoustic Emission Signal Analysis and Characterization

Investigation of predicting acoustic emission pulse durations in flat plate geometries was reported previously. A mathematical relationship which calculates the increased time duration of signals due to receiving energy from many ray paths was developed. Variables such as plate thickness, velocity, receiver diameter, distance from the source, etc., were included. The method was checked using actual emission sources as well as signals from small ultrasonic shear wave transducers. Measured signal lengths correlated well with calculated values.

The above relationship did not, however, define pulse shape. It is important to obtain information about signal shapes in order to optimize gating periods for data analysis methods such as emission rate and, ultimately, to assist in locating the emission source three dimensionally and describe the flaw in terms of acoustic emission. Variables and quantities essential for understanding pulse shape behavior are not easily determined. It appeared that such parameters as transducer transfer functions, etc., should be obtained before signal pulse shapes could eventually be determined. However, it was anticipated that most of these parameters need not be determined if the signal could be measured before any reflections within the plate had taken place. Then, with the same receiver and electronic system used, the signal could be measured again at a greater distance from the source so that it would include the effects of many reflections within the

plate. If the number of reflections within the plate, their reflection factor amplitudes and phases, and their differences in attenuation are known, it should be possible to predict the pulse shape at the greater distance.

To examine this initially, experiments with a shear wave artificial signal source were conducted, and the signal prior to its reflection was detected and recorded. This was then added to itself in amplitude, with phase shift information determined for the different ray paths reflected within the plate. The reflection factors and phase shifts were available using an EDPM program previously set up at BNW. Attenuation differences for different ray paths were found to be unimportant for these initial studies since the attenuation coefficient was only about 1.3 dB/ft at the 1.6 MHz signal frequency used.

In Figure 3.1a the original signal, measured sufficiently close to the source so that no reflections were present, is shown. This signal was also recorded on an X-Y recorder at the output of a sampling oscilloscope. This X-Y recording then permitted accurate determination of incremental amplitudes along the pulse. The pulse was subsequently added to itself as mentioned above. The total number of included reflections was calculated using information from the study discussed in the previous Quarterly Report. For the receiving transducer used, the calculation indicated that 46 reflections should be included. The addition of this many reflections by hand calculation appeared too time consuming and only the first twenty were actually added.

The measured and calculated signals respectively are shown in Figures 3.1b and 3.1c. Their similarity is apparent and it appears that such methods should be capable of predicting pulse shapes in other situations. The pulse

Neg 06900

1 μ sec

a) Original Measured Signal

5 μ sec

b) Measured Signal with 46 Reflections

Neg 0684728

5 μ sec

c) Calculated Signal with 20 Reflections

FIGURE 3.1. Measured and Calculated Signals

synthesis could be made easier and faster with either EDPM calculations or with properly designed electronic function generators to simulate initial unreflected signals and other associated delay features to permit addition of multiple functions representing reflected ray paths. Eventually then acoustic emission detection problems for a variety of situations encountered in practice could be mocked up in the laboratory. Prior to actual field applications, test parameters could then be investigated and optimized.

Task 2 - High Temperature Transducer Development

In keeping with indications from the wave propagation study that shear mode detection is preferable in acoustic emission work to optimize the detected signal, 1.5 to 2.5 MHz shear crystals are being used for laboratory test work. An encased version has been fabricated for testing with the case intended to alleviate problems of stray signal pickup by exposed connections on the bare crystals. This preliminary application of the shear sensor is intended to gain experience with consistency of response before investing in development of a general field application version.

Task 3 - System Analysis Study

No further work.

Task 4 - Monitor System Development

Reactor system noise and wave propagation studies have both shown the need to detect acoustic emission in the frequency range of 1 to 3 MHz to optimize reliability. A tuned, solid state preamplifier has been developed for this purpose. It has a bandwidth of about 600 KHz with the center frequency

adjustable through the range of 1 to 2.5 MHz. The noise level is 3.5 μ V (rms) referred to input and the gain is 150. Effective roll off on either side of the pass band is about 14 dB/octave. A tuned preamplifier has the distinct advantage of alleviating the problem of preamplifier saturation by high level, low frequency noise signals. It also minimizes the need for extra filtering.

Task 5 - Cooperative Work with Phillips Petroleum Company

No activity this period other than verbal liaison.

Task 6 - Application Engineering

A rupture test performed with 24 in. OD x 1.5 in. wall Type 316 stainless steel pipe has been monitored for acoustic emission. The test was conducted by Battelle-Columbus the week of December 16, 1968 as part of their program to study ductile pipe rupture. The specimen had a 6 in. long by 0.9 in. deep flaw milled in the wall, and it was pressurized by heating water in the vessel. Failure occurred at about 670 °F and 4050 psig. The remaining pipe wall at the bottom of the flaw cracked through about half the length of the flaw which produced very little distortion even in the near vicinity of the flaw. Six acoustic emission sensors were attached to the vessel wall at various distances from the flaw. The response from all six was recorded on magnetic tape and one channel was also monitored directly for emission rate and total emission. The profile of these curves correlated very well with perturbations in the curve generated from a crack-opening-displacement gauge installed on the flaw. Post-test analysis of the total acoustic emission data has been delayed by breakdown of the magnetic tape recorder and is now in progress. One inconsistency which has become apparent and is

so far unexplained is the fact that in some cases signals appear on some data channels but not on all of them. The need to determine the cause of this is obvious from the standpoint of emission detection reliability.

Acoustic emission is being applied to investigate potential slow crack growth by fatigue in a specific section of piping at one of the Hanford reactor plants. High, localized stress levels were detected on some 24 in. to 36 in. pipe laterals. One of these has been monitored for acoustic emission as the water pressure and flow were increased. At 113% of normal operating pressure, five emission signals were detected. They triangulated to an area of uncertainty about 3/4 in. in diameter. Subsequent examination of the area with ultrasonic techniques failed to show the flaw, thus confirming that it is very small. Figure 3.2 shows the monitor arrangement and samples of the detected emission signals. Continued surveillance of these pipe laterals (seven) by acoustic emission has been requested.

The acoustic emission monitor system installed on the Containment System Experiment simulator vessel at Hanford has been used during a blowdown test to monitor known flaws in the vessel heads. Data analysis did not show evidence of flaw movement. Confirmation that the emission monitor was functional was obtained from the fact that emission from normal loading of structural members was detected.

General

Three topical reports and two technical papers were produced this quarter to promote dissemination of information on acoustic emission.

A - TRANSDUCER LOCATIONS ON 24 in. TO 36 in. PIPE LATERAL

B - TYPICAL TRANSDUCER RESPONSE AT 465 PSIG PRESSURE

Neg 0690242-4

FIGURE 3.2. Pipe Fatigue Measurement by Acoustic Emission

Work Planned for Next Quarter

1. Initiate study of wave propagation mechanics in curved geometries.
2. Design a method of applying the shear sensor in the field.
3. Complete analysis of data obtained from heavy wall stainless steel pipe burst test.
4. Determine the maximum range of emission detection using a shear transducer.
5. Initiate study of ductile and brittle fracture process characteristics using 1.5 to 2.5 MHz shear wave sensing system.

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- The COL HEAT simulation system is fully capable of application to any reach or region for which planning for industrial discharges is required. Temperatures predicted by the model fall within error limits ordinarily associated with measurements by local instruments. It can be categorically stated from the outputs generated in the study that the system of computation known as COL HEAT can reproduce the thermal regimen of any river system in the work for which weather data and flows are known.
- The results indicate that because of the limitations imposed by State Water Quality Standards few, if any, sites will remain undeveloped by the year 2000 where flow through cooling may be utilized exclusively to cool effluents from plants with a capacity as large as 1000 MW.

Simulation of the thermal regimen of the Phelps County (Nebraska) Canal was initiated. The purpose of the study is to test whether regional weather and radiation data can be used in the simulation of hourly average canal temperatures. Detailed data of power production is available for a 100 MW steam plant, which utilized canal water for flow through condensor cooling.

Operation of thermographs installed under this program was continued during the period. Locations now being serviced include:

Illinois River below the Dresden	
Island Dam	River Mile 269
Illinois River at Starved Rock Dam	River Mile 233
Illinois River at Meridosia, Illinois	River Mile 80
Mississippi River at Alton, Illinois	at Dam 26

In order to complete the array for further testing of simulation capability as projected plants are brought on the

line, thermographs are being installed on the Mississippi below Hastings Dam near Minneapolis and in the vicinity of the Quad Cities. Independent reports of the data collected in this program will be made through the appropriate office of the U.S. Geological Survey. The location of instruments in support of the Ohio Basin study is still under study in order to determine what key locations require this type surveillance compared to routine measurements taken by other agencies. The AEC sponsored instruments are kept highly calibrated to assure a record with an accuracy of ± 0.20 °C. The COL HEAT model has been demonstrated to be able to reproduce temperature differences between two stations within 0.314 °C compared to a combined instrument and readout error for a differential measurement of 0.275 °C.^(1,2)

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A - TRANSDUCER LOCATIONS ON 24 in. TO 36 in. PIPE LATERAL

B - TYPICAL TRANSDUCER RESPONSE AT 465 PSIG PRESSURE

Neg 0690242-4

FIGURE 3.2. Pipe Fatigue Measurement by Acoustic Emission

4. ENVIRONMENTAL STUDIES

SIMULATION MODELING OF EXPECTED THERMAL GENERATION IN SELECTED RIVER SYSTEMS - R. T. Jaske and D. E. Peterson

Operations research with the COL HEAT thermal simulation model to investigate the effects of future nuclear power plants on temperatures of the Ohio River System is in the developmental stage. Solar radiation, weather, stream flow, and observed water temperatures have been obtained for the entire Ohio River main stem and also for several major tributary streams. In general the data are adequate for reconnaissance modeling of a major river system.

During the report period the following accomplishments were made:

- The report BNWL-999, A Test Simulation of Potential Effects of Thermal Power Plants on Streams in the Upper Mississippi River Basin, was completed. The report covered the potential impact of flow through cooling of nuclear generating plants up to the year 2000.
- With effective coordination of plant size and location, some river basins such as the Upper Mississippi offer relatively large opportunities for the use of direct discharge. The entire reach studied could probably accommodate the predicted growth of the power industry through 1980, and in some areas where development is quite slow through the year 1990.
- The study reveals that local control of policy decisions regarding mode of heat rejection does not necessarily reflect optimal development, and that in some cases decisions to use cooling towers in plants already under license or construction may have been premature.

- The COL HEAT simulation system is fully capable of application to any reach or region for which planning for industrial discharges is required. Temperatures predicted by the model fall within error limits ordinarily associated with measurements by local instruments. It can be categorically stated from the outputs generated in the study that the system of computation known as COL HEAT can reproduce the thermal regimen of any river system in the work for which weather data and flows are known.
- The results indicate that because of the limitations imposed by State Water Quality Standards few, if any, sites will remain undeveloped by the year 2000 where flow through cooling may be utilized exclusively to cool effluents from plants with a capacity as large as 1000 MW.

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5. FIXATION OF RADIOACTIVE RESIDUES

SPRAY SOLIDIFICATION

LABORATORY STUDIES

Corrosion Studies - R. F. Maness

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

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

ENGINEERING STUDIESDevelopment Spray Solidification Runs - J. D. Moore

A PW-4m phosphate feed suitable for spray solidification was developed and tested in the cold semiworks spray solidifier. It was necessary to dilute the feed to 585 liters/tonne to achieve adequate feed flow control. In two additional spray solidifier runs, ruthenium volatilization in the spray solidification process from PW-1 feed was shown to increase with feed acidity. At a feed acidity of zero, essentially no ruthenium was volatilized. Feed compositions for the three runs are listed in Table 5.1 and solidifier operating conditions and results are summarized in Table 5.2.

A. Run DSC-22

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

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

TABLE 5.1. *Feed Compositions for Development Spray Solidifier Runs (Basis: 378 liters/tonne concentration)*

Component	DSC-22	DSC-23	DSC-24
Waste Type	PW-4m	PW-1	PW-1
Fe ⁺³	0.124	0.955	0.946
Cr ⁺³	0.012	0.012	0.012
Ni ⁺²	0.008	0.005	0.005
Al ⁺³	0.001	0.001	0.001
ZrO ⁺²	0.143	0.065	0.065
Na ⁺	0.342	0.178	0.138
SiO ⁻²	0.010	0.010	0.010
PO ₃ ³	0.003	0.003	0.003
MoO ₄ ⁻²	0.13	0.065	0.065
Sr ⁺²	0.036	0.0155	0.0155
Ba ⁺²	0.041	0.0195	0.0195
Rb + Cs (used K ⁺)	0.092	0.042	0.042
(Y + RE) ⁺³	0.274	0.12	0.12
Ru ⁺⁴	0.0082	0.0064	0.016
Rh ⁺² (used Co ⁺²)	0.013	0.0074	0.0074
Ag ⁺	0.0016	0.0008	0.0008
Cd ⁺²	0.0025	0.0008	0.0008
TeO ₄ ⁻² (used SO ₄ ⁻²)	0.015	0.0064	0.0064
Pd ⁺² (used Ni ⁺²)	0.043	0.017	0.017
TcO ₄ ⁻²	0.031 (used MoO ₄ ⁻²)	0.014 (used MoO ₄ ⁻²)	0.014 (used Mn ⁺²)
NO ₃ ⁻	6.6	3.6	3.63
H ⁺³	4.8	0.022	0.022

Additives

	Batch #1	Batch #2	Batch #3	
Na ⁺	1.258	1.0	1.0	1.8 ^(a)
Fe ⁺³	1.2	--	--	--
PO ₄ ⁻³	2.46	0.69	1.015	1.34
H ⁺⁴	7.38	2.07	4.315	6.57
NO ₃ ⁻	3.6	--	1.27	2.55
OH ⁻	1.258	--	0.325	0.65
PO ₃ ⁻	--	1.0	0.675	0.35
				1.8 ^(a)

a. Added as solid to the melter.

TABLE 5.2. Summary of Development Spray Solidification Runs

	<u>DSC-22</u>	<u>DSC-23</u>	<u>DSC-24</u>
Feed type	PW-4m	PW-1	PW-1
Feed concentration, liters/tonne	470-585	454	378
Additives	Fe^{+3} , Na^{+} , PO_4^{-3}	Na^{+} , PO_3^{-} , PO_4^{-3}	NaPO_3 , solid frit
M ⁺ /P	2.75	2.9	3.0
Feed consumed, liters	125.5	105, part 1 80.5, part 2 74, part 3	181
Average feed, rate liters/hr	14	15.6	15
Calciner furnace temperature, °C	700	700	700
Calciner vacuum, in. of water	2-10	10	8
Atomizing gas	air	air	air
Atomizing gas flow, scfm	6	6	7
Filter blowback pressure, psig	30-40	40	32-40
Filter pressure drop, end of run, in. of water	3.75	5.75	5.5
Melter furnace temperature, °C	1100-1150	1150-1175	1150
Product receiver furnace temperature, °C	800	850	600
Product collected, kg	35.5	63	53.7
Product density, g/cm ³	3.5	3.6	3.3
Particulate DF (feed to con- densate)	1000	540	460
Product concentration, liters/tonne	50	31	34
Solid NaPO_3 added to melter, kg	0	0	34

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

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

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

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

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

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

B. Runs DSC-23 and 24

Runs DSC-23 and 24 were made to observe the effect of PW-1 feed acidity on volatilization of ruthenium from the feed in the spray solidification process. In DSC-23, simulated PW-1 feed with additives of Na^+ , PO_4^{-3} , and PO_3 was prepared in three batches with acidities ranging from 2.1 to 5.9M. Each batch was run for approximately 6 hr with identical spray solidifier operating conditions (see Table 5.2) while condensate, scrubber, calcine, and melt product samples were collected. In DSC-24, feed was made up to contain only 0.022M free acid as compared to a normal acidity of 3.7M (8.9M including melt-forming additives). The only melt-forming additive used in this run was sodium metaphosphate which was added as solid chips to the melter. Analyses of the condensate and caustic scrubber samples showed that ruthenium volatilization increased nearly linearly from less than 0.5 to 33 percent with feed acidity (see Figure 5.1). To verify the ruthenium data from liquid samples, noncondensable off-gases from the condenser were also treated during DSC-24. One SCFM (10 percent of total flow) from the noncondensable off-gas flow was continuously passed through a ruthenium trap consisting of formaldehyde-treated silica gel in series with a bed of small polyethylene rings. Both

*FIGURE 5.1. Ruthenium
Volatilization from PW-1
Feed in the Spray
Solidifier*

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

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

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

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

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

During the quarter, the eighteenth and nineteenth pilot plant demonstrations of high level radioactive waste solidification were successfully completed with spray solidification Runs SS-6 and SS-7. A total of 6,650,000 Ci of radioactivity were interfused in 129 liters of ceramic solid to produce a total internal heat generation rate of 23,500 W. Sulfate-free PW-1 waste with additives of LiOH, NaOH, and H₃PO₄ was processed. Tables 5.3 and 5.4 present a summary of data and feed compositions for each run.

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

* BNWL-923, 68-3

TABLE 5.3. Summary Data for WSEP Spray Solidifier Runs

Operational Data	SS-5	SS-6	SS-7
Solidifier Run/WSEP Run	5/17	6/18	7/19
Solidifier	Spray	Spray	Spray
Date	10/68	11/68	12/68
Operating Mode	A	A	A
Feed Type ^(a)	PW-1	PW-1	PW-1
Feed, liters/tonne	446	521	452
Pot Diameter, in.	8	8	8
Pot Material	304-L	304-L	304-L
Total Run Feed Time, hr	53.2	55.6	47
Average Feed Rate, iters/hr	16.7	17.7	17.9
Total Feed, liters		986	839
Total Feed, tonne equivalent	2.0	1.9	1.85
Feed: Solid Volume Ratio	13.3	15.6	13.1
Feed: Solid Volume Ratio at 378 liters/tonne Basis	11.4	11.3	11.0
Filter ΔP, in. of Water	6.0	6.0	6.0

Problems

- SS-5 Power lead to melter furnace lost insulation and began arcing; insulation replaced with 3 1/2 hr delay in run.
- SS-6 (1) Partial plugging of the calciner inlet to the melter required two shutdowns to remove calcine from this area.
(2) Calciner vibrator failed and was replaced.
- SS-7 Feed nozzle plugged halfway through the run due to inadequate feed flow measurement.

Filled Pot Data			
Total Curies in Feed to Pot ^(b)	2,500,000	3,030,000	3,620,000
Total Curies of Ru in Feed to Pot ^(b)	16,800	24,100	22,000
Solid in Pot, liters	66	65	64
Solid in Pot, kg ^(c)	214	207	206
Solid in Pot, liters/tonne	33	35	34
Heat in Pot, W ^(d)	8200	10,800	12,700
Equivalent Aging Time, years	1.1	0.73	0.55
ε Temp in Air-Cooled Furnace, °C	633	767	928
ε to Wall Δ Temp., °C	278	308	450
Effective Thermal Conductivity, W/(m ²) (°C/m) [Btu/(hr) (ft ²) (°F/ft)]	1.1 [0.64]	1.39 [0.79]	1.1 [0.64]
Welded Lid	Yes	Yes	Yes
Radiation Reading, R/hr at 6 in.	NA	5.1 x 10 ⁴	NA

NA Not Available.

a. See Table 5.2 for feed composition.

b. By feed analysis.

c. Calculated from lab density and rodded volume

d. By pot calorimetry data.

TABLE 5.4. PW-1 Feed Compositions for Spray Solidification Runs SS-5, SS-6, and SS-7

Component	SS-5		SS-6		SS-7	
	Molarity, M ^(a)	Lab Analysis, M ^(a)	Molarity, M ^(a)	Lab Analysis, M ^(a)	Molarity, M ^(a)	Lab Analysis, M ^(a)
H ⁺	--	5.27	--	6.3	--	
Fe ⁺³	0.793 ^(b)	0.75 ^(b)	0.797 ^(b)		0.792 ^(b)	0.69 ^(b)
Cr ⁺²	0.012		0.012		0.012	
Ni ⁺²	0.005		0.005		0.005	
Al ⁺³	0.137 ^(b)	0.19 ^(b)	0.133 ^(b)	0.105 ^(b)	0.137 ^(b)	
Na ⁺	0.138	0.67 ^(c)	0.138	0.596 ^(c)	0.138	0.58 ^(c)
U ⁺⁶	0.01		0.01		0.01	
NO ₂ -NO ₃ ⁻	7.5	6.4 ^(d)	7.5	6.7 ^(d)	7.5	4.7 ^(d)
SO ₄ ⁻²	--		--		--	
PO ₄ ⁻³	0.003		0.003		0.003	
SiO ₂ ⁻²	0.01		0.01		0.01	
Cs ⁺³ (added K ⁺)	0.0416		0.0416		0.0416	
Sr ⁺²	0.0155		0.0155		0.0155	
Ba ⁺²	0.0195		0.0195		0.0195	
(Y + RE) ⁺³	0.12		0.12		0.12	
ZrO ₂ ⁺²	0.065		0.065		0.065	
MoO ₄ ⁻² + Tc as MoO ₄ ⁻²	0.079		0.079		0.079	
Ru ⁺⁴	0.0032		0.0032	0.0039	0.0032	0.0039
Ru ⁺⁴ (added Fe ⁺³)	0.0288		0.0288		0.0288	
Rh ⁺² (added Co ⁺²)	0.0074		0.0074		0.0074	
Pd ⁺² (added Ni ⁺²)	0.0222		0.0222		0.0222	
Ag ⁺ + Cd ⁺² (added Cu ⁺²)	0.0016		0.0016		0.0016	
TeO ₄ ⁻² (added SO ₄ ⁻²)	0.0064		0.0064		0.0064	
Additives to Feed						
Li ⁺ (as hydroxide)	0.5		0.5		0.5	
Na ⁺ (as hydroxide)	0.5		0.5		0.5	
PO ₄ ⁻³ (as H ₃ PO ₄)	0.713	1.67	1.732	1.99	1.778	1.60
Final Feed						
Volume/tonne		466		521		452
Waste Equivalents						
EM _T ⁺	4.70		4.74		4.89	
EM _T ⁺ /P	2.75		2.75		2.75	
Sp Gr at 25 °C		1.36		1.32		1.31
Vol% Solias		~30		~20		~36
Total Radioactivity, Ci/liter ^(e)		2260		3070		4310
Radioruthenium, Ci/liter ^(e)		18.9		24.2		26.2
Total heat, W/liter ^(e)		9.27		11.3		15.3

a. Molarities are at 378 liters/tonne basis.

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

c. Includes sodium from additive.

d. Includes nitrate from additives.

e. By feed analysis.

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

Spray Solidification Run SS-5

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

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

The distribution of the feed radioactivity for Runs SS-5, SS-6, and SS-7 is given in Table 5.5. During Run SS-5, 70 percent of the ruthenium in the feed to the solidifier was volatilized

TABLE 5.5. Activity Distribution for Runs SS-5, SS-6, and SS-7 (Mode A Operation)*

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

NA Not Available

* BNWL-507, 67-2

a. $7^{-1} = 7 \times 10^{-1}$

from the solidifier, while only 0.06 percent of all other combined radioactive species were entrained. Figure 5.2 shows the ruthenium accumulation in the WSEP evaporator during Runs SS-5, 6, and 7.

FIGURE 5.2. *Ruthenium Accumulation in the WSEP Auxiliary Evaporator*

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

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

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

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

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

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

Spray Solidification Run SS-6

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

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

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

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

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

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

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

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

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

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

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

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

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

Spray Solidification Run SS-7

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

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

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

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

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

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

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

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

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

PHOSPHATE GLASS SOLIDIFICATION

LABORATORY STUDIES

Corrosion Studies - R. F. Maness

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

STUDIES ON AUXILIARIES TO SOLIDIFICATION SYSTEMS

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

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

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

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

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

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

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

Z = tube length, ft
R = tube diameter, ft
D = diffusivity, ft²/sec
V_{max} = centerline velocity, ft/sec.

DEVELOPMENT STUDIES OF SOLIDIFICATION PROCESSES

HEAT TRANSFER STUDIES

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

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

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

The following simplifying assumptions were used in the derivations:

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

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

Storage pot calculations without heaters have been reported in graphical form by W. V. DeMeir.* A figure included at that time on pot wall temperature estimates was in error and is presented here in its correct form as Figure 5.3.

FIGURE 5.3. Pot Wall Temperatures in Air

* BNWL-507, 67-2

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

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

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

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

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

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

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

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

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

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

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

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

Examples of Applications

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

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

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

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

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

Size of Molten Core in Cylindrical Storage Containers.

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

Description of a container of waste:

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

Thermal conductivity of container

wall:	9 Btu/(hr) (ft ²) (°F/ft) (15.6 W/(m ²) (°C/m)
Melting point of waste:	1380 °F (749 °C)
Wall thickness:	0.3 in.

From Figure 5.3 it is estimated that the container will lose heat to the surroundings at a rate of 60,000 Btu/hr (17,580 W) when the surface temperature of the container is 842 °F (450 °C).

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

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

Nomenclature

Variables.

- A = volumetric heat generation rate, Btu/(hr) (ft³)
- a = thickness of frozen waste, ft
- b = thickness of vessel wall, ft
- B = power of axial or central heater, Btu/hr
- c = constant of integration
- k = thermal conductivity, Btu/(hr) (ft²) (°F/ft)
- L = length of filled portion of storage vessel, ft
- r = radius of cylinder or sphere, ft
- T = temperature, °F
- π = 3.1415..., mathematical constant

Subscripts

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

Filling of Pots with Thermal Maximum Heat Content in Water -
J. L. McElroy

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

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

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

** BNWL-507, 67-2

of considerations given to filling a pot with a high quantity of fission product heat. For cylindrical containers filled to a depth of 6 ft, Figure 5.4 shows the heat generation rate required at various product thermal conductivities in order for the pot centerline to attain a 900 °C centerline temperature. Figure 5.5 gives the product frozen wall thickness as a function of remelt temperature. Figure 5.5 is for an 8 in. diameter pot containing waste with a heat rate content equal to the thermal maximum in water limit for a centerline temperature of 900 °C taken from Figure 5.4. As an example, suppose a thermal maximum in water pot (hypothetical 900 °C centerline) is desired for a product with a thermal conductivity of 0.7 Btu/(hr)(ft²)(°F/ft). From Figure 5.4 the heat generation rate required is 23 kW. If the product has a melting point of 700 °C, the frozen wall thickness during water storage would be 2.0 in. (Figure 5.5). If the pot lost cooling water but had the benefit of natural convection cooling, the frozen wall thickness would decrease to about 1/2 in. Obviously, the later situation would give some cause for concern and some knowledge about the corrosiveness of the melt and the integrity of the frozen wall would be essential.

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

FIGURE 5.4. Heat Required in a Cylindrical Pot to Attain a 900 °C Centerline Temperature

FIGURE 5.5. Product Frozen Wall Thickness for 8-inch Thermal Maximum Pots

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

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

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

COMPOSITION OF
PG-4 SULFATE CONDENSATE

Fe	0.0089M
Al	0.018
Na	0.017
H ₃ PO ₄	0.097
HNO ₃	0.27
H ₂ SO ₄	0.40

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

Unique features of the flowsheet were:

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

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

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

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

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

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

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

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

The condensate was collected in four increments. Experimental results are shown in Table 5.6. The ruthenium distribution between condensate increments in Runs SD-18 and 20, made under identical conditions, differed markedly. The difference is thought to be due to irreproducibility of plating and flushing on the condensing surfaces. The conclusion is that very little of the volatilized RuO_4 condenses or is soluble in the

TABLE 5.6. Sulfate Condensate Distillation: Effects of Boilup Rate

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

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

b. Trapped on silica gel bed.

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

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

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

Development Studies - W. T. McKean

Cold runs with the Distillation Demonstration Unit (DDU), Figure 5.6, have been completed using acid mixtures containing 0.19M H_3PO_4 and 0.0067M Ru with sulfuric acid concentrations in the range of 0.5 to 5.45M and nitric acid from 0.455 to 2M. The runs are summarized in Table 5.7.

The following summarizes the outstanding results:

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

FIGURE 5.6. Sulfate Condensate Distillation Demonstration Unit (DDU)

TABLE 5.7. Summary of Distillation Demonstration Unit

Run No.	Acid Concentration, M		Boilup, gal/hr	Pressure, mm Hg	DF		Comments
	HNO_3	H_2SO_4			HNO_3	H_2SO_4	
A	1	5.45	2.2	50	855	1559	EDL-102 Cold Test
F	1	5.45	1.5	25	<3000	1017	
G	1	5.45	1.3	100	73000	1392	
B	1	1	1.7	50	44	5500	
H	1	1	1.9	25	1162	7167	
C	1	0.5	2.1	50	1160	11200	
I	1	0.5	2.2	50	31	309	Analysis not complete
D	0.455	0.5	2.3	50	78	252	
J	0.455	0.5	1.7	30	326	2688	
E	2.0	1.0	1.9	50	76	64	
M	2.0	1.0		25			Not complete

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

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

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

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

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

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

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

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

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

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

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

Water boilup tests and three cold runs have been made in C-Cell. The feed composition of the cold runs was identical to the out-of-cell cold Runs D and J listed in Table 5.7. These runs were duplicates of cold Runs D and J and were made to determine how closely DF values can be reproduced and to find if there is a trend toward lower DF values as subsequent runs are made. Such a trend would indicate ruthenium retention and carry over from early runs to later runs. Sample analysis for these runs is in progress.

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

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

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

EVALUATION OF SOLIDIFIED WASTE PRODUCTSNONRADIOACTIVE LABORATORY STUDIESStudy of High Temperature Storage of Solidified Waste Products Containing Sulfate - D. W. Brown and J. E. Mendel

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

The release of SO_3 from simulated spray solidifier PW-2 product was measured as a function of time over the temperature range 550 to 1000 °C. The release of SO_3 was followed gravimetrically, assuming all weight lost was SO_3 . The results, given in Figure 5.7, show an appreciable rate of sulfate decomposition even below the melting point of the product (~725 °C).

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

FIGURE 5.7. Sulfate Loss from Spray Solidifier PW-2 Product Held in Air

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

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

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

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

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

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

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

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

Thermal Conductivity

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

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

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

where,

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

α = thermal diffusivity, cm²/sec

ρ = density, g/cm³

C_p = heat capacity, cal/(g) (°C).

Thermal Diffusivity

Thermal diffusivity was measured on all sixteen samples at several temperatures between 30 and 400 °C. Values are reported in Table 5.8. In general, the samples seem to fall into two types. One with a low diffusivity which changes only slightly with temperature, and the other with a somewhat higher value which decreases sharply with increasing temperatures. Most of the more crystalline samples fall into the latter group.

Heat Capacity

The heat capacity of three samples has been determined using a Perkin-Elmer differential Scanning Calorimeter. Of the three samples, one was from the group having a high temperature sensitive thermal diffusivity and the other two were from the low, less temperature-sensitive group. The heat capacity and the thermal conductivity of these three samples are recorded in Table 5.8.

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

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

Product Characterization

Several pot calorimetry values were obtained in B-Cell.

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

All heat values are related to the run date.

TABLE 5.8. *Characterization of PW-2 Phosphate Glass Samples*

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

TABLE 5.8. Characterization of PW-2 Phosphate Glass Samples (contd)

Sample	A	B	C	D	$10^{-4} \alpha, \text{cm}^2/\text{sec}$	Temp, °C	$C_p, \text{cal}/(\text{g})(^\circ\text{C})$	Temp, °C	$10^{-3} k, \text{cal}/(\text{cm})(^\circ\text{C})$	Temp, °C	$\rho, \text{g}/\text{cm}^3$
10	1200	1	30	600	44	28	.162	50-100	1.96	75	2.88
					44	29	.166	100-150	1.91	125	
					42	75	.171	150-200	1.91	175	
					39	126	.199	200-250	2.06	225	
					37	200	.212	250-300	2.13	275	
					35	254	.226	300-350	2.15	325	
					33	325	.282	350-400	2.59	375	
					33	335					
					33	400					
11	1000	25	100	100	28	32					2.89
					28	59					
					28	75					
					27	131					
					27	200					
					23	255					
					28	266					
					29	321					
					32	347					
					25	362					
12					29	32	.181	50-100	1.53	75	2.91
					27	35	.181	100-150	1.48	125	
					29	57	.204	150-200	1.66	175	
					29	75	.222	200-250	1.74	225	
					27	200	.210	250-300	1.65	275	
					27	215	.251	300-350	1.97	325	
					27	250	.257	350-400	1.94	374	
					27	321					
					26	400					
13	1200	25	30	100	35	33					2.90
					31	52					
					31	75					
					31	77					
					35	112					
					30	159					
					34	165					
					34	192					
					36	200					
					34	219					
14	1200	1	100	100	29	28					2.92
					29	30					
					26	123					
					21	213					
					26	250					
					26	235					
					25	352					
					25	400					
15	1200	25	30	600	30	34					2.92
					30	35					
					31	85					
					30	92					
					29	97					
					29	125					
					29	206					
					28	252					
					28	325					
					27	400					
16	1000	25	100	600	49	28					2.93
					44	80					
					41	125					
					37	208					
					35	250					
					34	325					
					32	390					
					29	400					
17	1200	25	100	100	29	32					2.91
					27	86					
					26	125					
					26	144					
					26	204					
					26	250					
					26	257					
					23	325					
					25	400					

Note:

A = Melt Temperature.

B = Hours in Melt.

C = Cooling Rate °C/hr.

D = Eight Week Storage Temperature.

6. HEAVY SECTION STEEL TECHNOLOGY PROGRAM

FRACTURE STUDIES OF PRESSURE VESSEL STEELS - C. W. Hunter,
J. A. Williams and C. L. Hellerich

HSST Program: Irradiation Effects

The objective of this program is to obtain a quantitative engineering evaluation of the effects of irradiation and reactor environment on the plane strain fracture toughness (K_{I_C}) of heavy-section pressure vessel steels. The past study has centered on the HSST program plate 01 (A 533-B) using the Double Cantilever Beam (DCB) specimen.^(1,2) This investigation delineated the valid plane strain measuring capacity of the DCB specimen, defined the K_{I_C} between -160 °C and -80 °C, and defined the K_{I_C} profile through the 12 in. plate.

In the current work, 1-T Compact K_{I_C} specimens are being utilized in place of the DCB specimen. The specific effort in progress now is the determination of the effect of irradiation on the K_{I_C} of A 533-B base material and weldment. Work on assessing the deleterious effect of strain aging (during neutron exposure) is in the planning stage. Programs on warm prestressing and on fatigue have been proposed but are not in progress. Specimens for the base material and strain-aging studies have been machined from the 12 in. HSST program plate 02. The specimen orientation is RW and specimens were taken from the 1/4-t through 3/4-t positions.

The weldment being studied is the HSST program weldment section 51A that had been submerged arc welded from plate 01. It is 30 in. long by 12 in. thick. Metallography of the weldment has been completed in preparation for locating the fracture mechanics specimens. The goal of the weldment study

is to determine the K_{IC} gradient from the base material, through the HAZ and into the weld metal. Both 1-t compact K_{IC} (WOL) and 1/2 in. DCB specimens are being employed in the orientations shown in Figure 6.1 to assess the two types of failure modes. The 30 in. of weldment will permit K_{IC} determinations for one unirradiated and one irradiated condition.

The desired irradiation fluence levels are 1×10^{19} n/cm² at 232 °C (450 °F) and 287 °C (550 °F) and 8×10^{19} n/cm² at 287 °C (550 °F). The first irradiation capsule contains 10 compact K_{IC} specimens and is to be charged into the M-3 hot water loop of the Idaho Nuclear Experimental Test Reactor (ETR) in January 1969. The capsule will also accommodate 30 tensile specimens. These specimens are for the base material study and will be irradiated at 287 °C (550 °F). Specimen temperature control will be maintained by gamma heating and the coolant flow rate. Each specimen temperature will be monitored, as will the fluence at the notch root. Plans are being formulated to modify the heat exchanges in the M-3 loop and to develop a capsule for the reflector positions in the ETR so that the 232 °C irradiations may be achieved.

Irradiation Embrittlement Sensitivity

This task is a cooperative effort with NRL to determine the possible causes of variable radiation embrittlement in pressure vessel steels and weldments. During the past several years, NRL has experimentally demonstrated that Cu, P, and V enhance embrittlement at fast fluence levels of approximately 3×10^{19} n/cm².⁽³⁾ This embrittlement has been reflected in an elevated C_v 30 ft-lb transition temperature and a depressed upper shelf energy. The BNW portion of this effort is to determine the nature of the irradiation damage using fractography, optical, and transmission electron microscopy, and microprobe analysis.

TEST METHODS

WOL

1. MACROETCH
2. LOCATE AND MACHINE NOTCH

DCB

1. AVOIDS LOCATION DIFFICULTY
2. FEWER SPECIMENS

6.3

3. DIFFICULT TO IDENTIFY ABOVE STRUCTURES
4. CURVED CRACK REMAINS IN MATERIAL
3. AVERAGE CRACK EXTENSION 0.5"
4. GRADIENT DETERMINED BY CHANGE

FIGURE 6.1. Test Methods Being Employed in the Determination of the K_{Ic} Gradient Through the Weld

BNWL-1009

The materials being studied are:

- (1) A 302-B, 6 in. reference plate,
- (2) A 302-B, 1/2 in. plates from split heats with and without residual elements,
- (3) A 543, 4 in. plate, which was embrittlement sensitive,
- (4) A 543, 1 in. plate, which was embrittlement insensitive, and
- (5) A 543, 4 in. weldment, which was very embrittlement sensitive.

Except for (1) above in the unirradiated condition, only several broken Charpy halves were available from NRL for each material. For fractography, it was, therefore, necessary to select temperatures for refracture that were characteristic of material behavior. The temperatures selected were those on the lower or upper shelves for the respective materials and conditions. The philosophy behind this choice was to achieve a meaningful way to compare the surfaces. Fractures on the lower and upper shelves correspond to plane strain and plane stress conditions respectively, and therefore, any differences in the fracture surfaces are attributable to material properties. Fractures positioned in between depend on an unknown function of constraint, geometry, material, and temperature. The upper shelf examination of the A 543 4-in. weldment will be especially fruitful since it exhibited such a low upper-shelf energy.

Enough unirradiated A 302-B 6-in. reference material was available to characterize the fracture surfaces throughout the Charpy transition temperature range. Figure 6.2 is the Charpy impact energy curve and Figures 6.3 through 6.11 are typical fracture surfaces as revealed by scanning electron microscopy. A comparison of Figures 6.3 and 6.4 depicts the effect of constraint on the fracture mode and the fracture initiation site. The stereo fractograph in Figure 6.5 illustrates typical

FIGURE 6.2. Charpy Impact Energy for A 302-B 6-in. Reference Material in the Unirradiated Condition

5000X

FIGURE 6.3. Plastic Dimpling in the Shear Lip at the Base of the Notch. [The dimples point toward the direction from which the fracture initiated, i.e. internally. The arrow indicates the macroscopic fracture direction. Impact fracture of A 302-B at -40°C (-40°F). Scanning electron micrograph.]

2000X

FIGURE 6.4. Cleavage at the Edge of the Shear Lip Just Above Figure 6.3. [The river patterns point toward the notch (down) indicating the fracture initiated as cleavage internally and then turned back towards the notch. Impact fracture of A 302-B at -40°C . Scanning electron micrograph.]

1000X

FIGURE 6.5. *Typical Region of Cleavage. (Note the extremes of topography and occurrence of tear ridges. The river markings indicate a very discontinuous fracture propagation. Impact fracture of A 302-B at -40 °C. Scanning electron stereo micrograph.)*

6.8

1000X

FIGURE 6.6. *Cleavage Fracture Exhibiting Cleavage Cracks Normal to the Main Facet. [This is consistent with (100) cleavage plane orientation. Note that the cracking occurred secondary to the main fracture as evidenced by the continuation of river markings across the crack. Impact fracture of A 302-B at -40 °C. Scanning electron stereo micrograph.]*

BNWL-1009

5000X

FIGURE 6.7. *Typical Plastic Dimpling at the Notch Root. [The elongation of the dimples resulted from the extremes in topography. Impact fracture of A 302-B at 26 °C (80 °F). Scanning electron stereo micrograph.]*

6.10

2000X

FIGURE 6.8. *Cleavage Facet Just Internal from the Plastic Shear Lip at the Notch Root. (River markings pointing towards the notch root indicate the initiation was internal. Impact fracture of A 302-B at 26 °C. Scanning electron stereo fractograph.)*

BNWL-1009

300X

FIGURE 6.9. *Web-like Topography Resulting from a Network of Inclusions in a Region Which Fractured by Plastic Dimpling. (The location is in the shear lip on the side of the specimen. Impact fracture of A 302-B at 26 °C. Scanning electron stereo micrograph.)*

3000X

FIGURE 6.10. Detailed View of the Web-like Topography in Figure 6.9. (The partical morphology is indicative of Mn5. Each inclusion was a site of local initiation and dimpled tear ridges formed in between. Impact fracture of A 302-B at 26 °C. Scanning electron micrograph.)

300X

FIGURE 6.11. Detail of a Hole Resulting from Local Fracture Initiation at Each Inclusion. (Impact fracture of A 302-B at 49 °C (120 °F). Scanning electron fractograph.)

(quasi) cleavage. The plastic dimpling in Figure 6.7 was more extensive at 26 °C (80 °F), but still about 40% of the specimen center was cleavage, as evidenced in Figure 6.8.

Special attention has been directed toward the influence of inclusions on fracture. In an area of plastic dimpling, inclusions serve as a site of local fracture initiation and, thus, form a hole or pattern, as shown in Figures 6.9 through 6.11. The original inclusion network is reflected in the fracture topography. No inclusions were revealed on cleavage facets.

This fractographic study of the unirradiated A 302-B 6-in. reference plate is serving as base information with which the other specimens are being compared. A thinning technique has also been developed so that transmission electron microscopy may be performed.

Evaluation of Detrimental Metallurgical Structure in the CSE Vessel

Ultrasonic inspection had indicated flaws near a nozzle of the 4 in. thick Containment Systems Experiment vessel. Core samples taken from the flawed regions were tensile tested and then metallographically and fractographically examined. The metallographic examination revealed that the flaws were parallel to the plate surface and were in base metal below the HAZ weld repair. Therefore, the flaws were a portion of those that were to have been repaired by welding. Electron microprobe examination of a section through the flaws indicated the presence of calcium and aluminum (oxide) in the flaws. These results are shown in Figure 6.12.

350X

Silicon

350X

Aluminum

250X

Photomicrograph

350X

Calcium

Photomicrograph and Electron Microprobe Results on a Cross Section Through the Flaws. (Note the association of calcium and aluminum with flaws, while only one spot contained silicon.)

FIGURE 6.12.

Tensile tests demonstrated that the metal, per se, had adequate strength and ductility. However, the flaws did separate into a crack at low strain levels (approximately the strain at metal yield). The fracture surfaces were examined by scanning electron microscopy and electron microprobe analysis. Figure 6.13 is a flawed region and Figure 6.14 shows the particles of the flaw. The electron microprobe results shown in Figure 6.15 revealed that the particles were calcium and aluminum (oxide). No evidence of other substances contributory to the flaws were found. Except for the flaws, the fracture was entirely by plastic dimpling.

This study demonstrated the merit of the coordinated application of scanning electron microscopy and electron microprobe analysis. Especially in the area of fracture analysis, such an approach is the only means of attributing fracture behavior to specific structure or microconstituent in a material.

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100X

FIGURE 6.13. *Flawed Region on the Fracture Surface. [The flaw (central bright areas) resulted from numerous inclusions. See Figure 6.14 for a detailed view of inclusions. The surrounding fracture was plastic dimpling. Scanning electron micrograph.]*

3000X

FIGURE 6.14. *Particles Which Comprised the Flaw in Figure 6.13. (See Figure 6.15 for microprobe analysis. Scanning electron micrograph.)*

1000X
Results from Electron Microprobe Scan for Aluminum and Calcium on the Flaw in Figures 6.13 and 6.14. (No indication of silicon, manganese, or sulphur was found.)

FIGURE 6.15.

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