NUCLEAR SAFETY QUARTERLY REPORT
AUGUST, SEPTEMBER, OCTOBER, 1968
FOR
NUCLEAR SAFETY BRANCH
OF
USAEC DIVISION OF REACTOR
DEVELOPMENT AND TECHNOLOGY

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FOREWORD

This report is the seventh 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.

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1. SUMMARY

ENGINEERED SAFEGUARD STUDIES

Containment Systems Experiment (CSE)

Two large scale experiments on the effectiveness of containment spray systems for reducing airborne fission product concentration were performed. In addition, the results of an earlier experiment on natural removal processes were evaluated. The initial containment atmosphere in all three runs was a steam and air mixture at about 250 °F and 35 psig. The aerosol included iodine, methyl iodide, cesium, and fumes from molten Zircaloy-clad UO_2 .

Results of the natural transport Run A-5 confirmed results from earlier similar runs (A-1 and A-2) at lower temperatures that aerosols and vapors released into a steam and air containment atmosphere were rapidly mixed and that the airborne aerosol concentration within any single compartment is quite uniform. Experimental data for removal of elemental iodine were in excellent agreement with a predictive model based on natural convection and the heat-mass transfer analogy. All aerosol components except methyl iodide were effectively removed by natural processes until airborne concentrations were less than 0.1% of initial values.

Initial results on fission product leakage rate at a penetration were obtained for a nominal containment vessel leakage rate of 0.3 wt%/day. Leakage of iodine and cesium showed an initial depletion, followed by enrichment at later times as compared with the volumetric leak rate of noncondensable gas. The overall decontamination factor for the first 5 hr of the run was about unity.

The first spray test, Run A-6, using a spray solution containing 3000 ppm boron as H_3BO_3 with NaOH to give a pH of 9.5, gave results quite similar to Run A-4 with an air

atmosphere and the same spray solution. During the first 10 min spray period, the effective removal half life of elemental iodine was 1.9 min, as compared with a predicted half life of 1.76 min based on absorption theory with gas phase diffusion limiting. As in previous spray runs, the second 30 min spray did not maintain this rapid removal rate. After a final 1 hr recirculation spray period, the overall decontamination factor for elemental iodine was about 2000.

The next spray test, Run A-7, evaluated iodine removal by a boric acid solution at pH 4.8 with all other conditions as for Run A-6. Only preliminary results from a single "thief" sample location are available at report time, but from these it appears that elemental iodine was removed with about a 2.5 min half life during the first spray period, and that the total decontamination factor for elemental iodine was about 1000.

Run A-7 also included a fourth spray period to evaluate methyl iodide removal by a basic sodium thiosulfate spray solution. Preliminary results indicate a removal half life of about 130 min.

Removal of particles by containment sprays has been evaluated in Runs A-3, A-4, and A-6. Theoretical treatment of removal by impaction, interception, and diffusion processes for different particle sizes and densities and for spray drop size in the range of interest has been compared with the experimental results. The particle sizes predicted to give the observed removal half times are in reasonable agreement with the particle sizes measured by cascade impactor.

Preparations for resuming experimental blowdowns continued. Cold mechanical loading tests were started as part of the investigation of the propagation of the defect indication in the bottom head of the CSE reactor simulator vessel.

The FLASH2 computer code for analysis of loss-of-coolant accidents was applied to CSE bottom nozzle blowdown experiments

with 2, 4, and 8 in. nominal break sizes. The results indicate that the selection or estimation of the proper effective orifice coefficient for the two-phase critical flow model in the code is a problem in application of the code.

Analysis of the liquid level in the reactor simulator vessel during blowdown has shown that the rate of liquid level decrease is relatively slower for larger break sizes when plotted as a function of a dimensional time parameter to correct for outlet area and initial liquid mass differences. The relatively slower rate of level decrease is only partly accounted for by the lower mass velocity for larger break sizes.

Particle Transport to Surfaces by Condensing Steam

Analog computer routines were designed to solve boundary layer equations which yield velocity, temperature, and mass profiles near a vertical surface upon which steam is condensing. The solutions obtained for mass transfer were shown to give almost identical results for condensing steam as equations generally accepted for heat transfer. This agreement reinforces the analog solutions for steam flux to a wall. Interfaced analog-digital computers (hybrid) are being used to solve the problem of particle transport in condensing steam. Experimental design was nearly complete, but until further funding can be obtained, the experimental phase will be postponed.

PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

Crack Detection in Pressure Piping by Acoustic Emission

Basic wave propagation study for the purpose of improving acoustic emission detection techniques has produced, to date, evidence that shear wave detection offers distinct advantages in signal-to-noise ratio and signal resolution.

High cycle, tension-tension fatigue testing of 304L Stainless Steel has shown the onset of acoustic emission activity at about 20% of the total number of cycles required to produce a visible surface crack.

Design has been completed for an environmental test of transducer crystal materials to determine the combined effect of high temperature and nuclear radiation.

A circuit to provide a continuous record of accumulated number of acoustic emission events has been satisfactorily tested. Testing has been performed to demonstrate detection of acoustic emission in the presence of turbulent flow and cavitation hydraulic noise. The results show that an acoustic emission sensing system operating in the 1.5 to 2.5 MHz frequency range with shear sensors is essentially insensitive to either turbulent flow or cavitation noise. At the same time, this system will detect acoustic emission signals clearly at a distance of at least three feet from the signal source.

An acoustic emission monitor system installed on the Containment Systems Experiment simulator vessel has been operated in conjunction with integrity evaluation tests on the vessel with satisfactory results.

FIXATION OF RADIOACTIVE RESIDUES

Spray solidification Run SS-4 was completed with 500 liters of sulfate containing PW-2 waste being reduced to 56 liters of ceramic solid in 40.5 hr. The 1,300,000 Ci of fission product activity interfused in the solidified waste produced an internal power generation of about 5400 W. Operation of the spray solidification systems was quite smooth, and all the equipment performed satisfactorily. The temporary loss of the mount for the calciner vibrator resulted in an interruption of the run, but a new floating-type vibrator was placed on the calciner and the run was successfully completed.

A new method has been developed to control the discharge of melt from the spray solidifier melter. The melt in the discharge valves is solidified by an air stream. To discharge the melt, the cooling air is turned off and the heat from the melter

furnace thaws the material in the freeze valve. This method of melt discharge was successfully used during spray solidification Run SS-5. During SS-5, 887 liters of sulfate free PW-1 waste, containing 2,000,000 Ci of radioactivity, was successfully reduced to 66 liters of ceramic solid in 53.2 hr. All spray solidifier systems performed satisfactorily.

Investigation of PW-4m feed formulations indicates that a phosphate rather than a borosilicate flowsheet will be favored for processing PW-4m in the spray solidifier. Among reasons for this are: rare earth precipitates form in the PW-4m borosilicate melts and extreme difficulty is encountered in preparing and handling the borosilicate feeds.

In a PW-4m run with the development spray calciner, a PW-4m flowsheet was found suitable for spray solidification when the borate flux was added separately as a solid to the melter rather than including it in the calciner feed.

The vacuum distillation for treating sulfate condensate resulting from phosphate glass solidification was tested with a simulated solution and tracer ruthenium. The overall decontamination factor was greater than 5×10^3 .

HEAVY SECTION STEEL TECHNOLOGY PROGRAM

Irradiation Damage to Heavy Section Vessel Steels

The plane strain fracture toughness, K_{IC} , is determined for ASTM A 533 B pressure vessel steel between -160 °C and -80 °C using various size Double Cantilever Beam (DCB) fracture specimens. Initial validity criteria for fracture testing low yield strength steels using the DCB specimen are established. Additional studies of the DCB specimen are indicated. Initial designs of fracture specimens irradiation capsules have been completed.

2. ENGINEERED SAFEGUARD STUDIES

CONTAINMENT SYSTEMS EXPERIMENT - G. J. Rogers

Large-Scale Fission Product Transport Experiments - L. F. Coleman, R. K. Hilliard, C. E. Linderoth, J. D. McCormack, M. E. Witherspoon, and G. J. Rogers

Two fission product transport experiments, Runs A-6 and A-7, were performed in the large CSE containment vessel during this report period. Containment spray systems were operated during both of these experiments. In addition, results of Run A-5, a natural transport test made near the end of the previous quarter, were evaluated.

CSE Run A-5

The primary objective of Run A-5 was to obtain experimental data on fission product behavior in the containment vessel in an environment similar to that expected after a Loss-of-Coolant Accident to a large PWR, and to compare the observed behavior with predictions based on mathematical models. To achieve the latter objective, several secondary objectives were included: (1) rate of mixing of FP simulant material in the atmosphere, (2) characterization of the fluid dynamics (temperature and velocity gradients, steam-air ratios, and steam condensation rates), (3) measurement of FP aerosol properties in the containment environment, (4) determination of the FP leakage rate through selected leak points, and (5) visual observations of the atmosphere during the test.

Test Conditions. The conditions for Run A-5 were the same as used in an earlier experiment, Run A-2.⁽¹⁾ with two differences. In the former test the containment vessel walls were not insulated; hence heat transfer and steam condensation rates were relatively high. Prior to Run A-5 all exterior surfaces

had been covered with sufficient insulation to reduce the overall heat transfer coefficient by a factor of ten. The temperature and pressure in the main vapor space during Run A-2 were 185 °F, 23.2 psia; in Run A-5 they were 253 °F, 47.0 psia.

The important test conditions are listed in Table 2.1. Figure 2.1 is a schematic diagram of the test arrangement used in Run A-5.

Simulant Aerosol Release. The simulant fission product materials were generated in a laboratory adjacent to the containment vessel and injected into the vessel as shown in Figure 2.1. Prior to aerosol injection the containment vessel was sealed with room temperature air at barometric pressure and brought to the desired test temperature by adding steam. The vessel had been at thermal equilibrium for about 35 hr when the aerosol was released.

The duration of the release was 10 min, short enough to be considered instantaneous for times longer than the release time. Zero time was defined as the time iodine and cesium release started. Methyl iodide was added by a separate injection line so that it did not pass through the high temperature furnaces. The initial iodine concentration in the containment atmosphere was in the range considered to be the maximum possible in the event of a complete core meltdown in a large power reactor.

Mixing in the Gas Phase. The mass concentration at various locations in the gas phase was measured by taking Maypack samples at all locations simultaneously for 3 min each. As in previous tests,⁽¹⁾ the concentration in the main room and drywell was found to be well mixed. The standard deviation from the mean of twelve locations was $\pm 20\%$ (68% confidence). Though no samples were taken during the 10 min aerosol release period, the first sample was started 2 min after release had terminated and was found to be as well mixed as in later samples.

TABLE 2.1. Summary of Test Conditions,
Runs A-5, A-6, and A-7

	A-5	A-6	A-7
Date of Test	7-31-68	9-24-68	10-15-68
Vessel	Main CSE vessel plus open drywell. Wet wells sealed off.	Same as A-5	Main CSE vessel plus open drywell plus 4 wetwells
Previous exposure to steam, hr	586	730	850
Avg. temp., main room, °F	253 ± 3	250 ± 3 ^(e)	250 ± 2 ^(f)
Avg. temp., middle room, °F	233	225	231
Avg. temp., lower room, °F	233	210	200
Avg. total pressure, psia	47.0	44.0 ^(e)	50.0 ^(f)
Avg. steam makeup during steady state, lb/hr	300	350	N.A. ^(g)
Condensing steam flux on main vessel walls, lb/hr ft ²	0.030	0.035	N.A.
Aerosol release duration, min.			
(Cg) ₀ , calc'd initial conc. in main room, mg/m ³	10	10	10
Total iodine	140	N.A.	N.A.
Methyl iodide	1.6	N.A.	N.A.
Cesium	1.8	N.A.	N.A.
Uranium-Zircaloy	~3	~3	~3
No. of spray nozzles ^(a)	None	12	12
Spraying pressure, psid	-	40	40
Flow rate, gal/min	-	49.1	49
Spray drop diam. ^(a) MMD, μ	-	1210	1210
Fall height to deck ft	-	33.8	33.8
to drywell, ft	-	50.5	50.5
% Upper volume sprayed	-	80	80
Spray solution	-	(b)	(c,d)

(a) *Spraying Systems Co. Catalog No. 3/4 7G3*

(b) *3000 ppm boron in NaOH solution, pH = 9.5*

(c) *3000 ppm boron in demin. water, pH = 4.8*

(d) *A fourth spray period used 1 wt% sodium thiosulfate, 3000 boron, pH = 9.3.*

(e) *Conditions at start of first spray. Temperature decreased to 200 °F and pressure to 30.0 psia at end of second spray.*

(f) *Conditions at start of first spray. Temperature decreased to 200 °F and pressure to 35.0 psia at end of second spray.*

(g) *N.A. signifies results not available at time of writing.*

①. Wall Condensate Collector	⑩. Viewing Window
②. Drywell Lid	⑪. "Thief" Sampler
③. Meter	⑫. Circumferential Condensate Collector
④. Drywell	⑬. Steam Jet Injector
⑤. Wetwell (Sealed Off)	⑭. Superheated Steam
⑥. Liquid Samplers	⑮. Fission Product Aerosol
⑦. Maypack Cluster (15 Total)	⑯. Makeup Steam
⑧. Main Containment Vessel	⑰. Middle Room
⑨. 1" Thermal Insulation	⑱. Lower Room
- $$k = 0.026 \left(\frac{\text{Btu-ft}}{\text{hr. } ^\circ\text{F ft}^2} \right)$$

FIGURE 2.1 Schematic Drawing of Containment Arrangement used in CSE Run A-5

Concentrations in the middle and lower rooms were much lower than in the main room at early times, but increased with time as aerosol material diffused into these lower regions. At long times the concentration in the lower rooms approximately equaled that in the main room.

The conclusion is that concentrations can be considered homogeneous within the bulk gas space in any individual room, but may differ appreciably between rooms.

Gas Phase Concentration Time Dependence. Gas phase concentrations are plotted as a function of time for the several mass transport species studied in Figures 2.2 through 2.7. As mentioned, the gas space in the main room plus drywell could be considered homogeneous, so only the average concentration is plotted, with the standard deviation from the mean being indicated.

All experimental points are corrected for background error by subtracting the value measured by a "blank" Maypack at each location. The points for elemental iodine and particulate-associated iodine have been further corrected for elemental iodine adsorption on the Maypack filter media. Both of these corrections are inconsequential for early times and become significant only after several hours containment time.

The results shown by these figures are consistent with those of earlier tests in the CSE and, as will be shown in a later section, in remarkable agreement with predictions based on natural convection heat transfer theory.

Elemental Iodine concentration in the main gas space (Figure 2.2) decreased exponentially with a half life of 13.5 min until it was about 0.1% of its initial value. This rapid decrease is attributed to transfer from gas to surfaces, with the rate being gas film diffusion controlled. After about

FIGURE 2.2 Elemental Iodine Concentration in Gas Phase - Run A-5

FIGURE 2.3 Particulate Iodine Concentration in Gas Phase - Run A-5

FIGURE 2.4 Methyl Iodide Concentration in Gas Phase - Run A-5

FIGURE 2.5 Total Iodine Concentration in the Gas Phase - Run A-5

FIGURE 2.6 Cesium Concentration in the Gas Phase - Run A-5

FIGURE 2.7 Uranium Concentration in Gas Phase - Run A-5

two hours the liquid (steam condensate) became saturated and further reduction in gas phase concentration occurred only slowly (24 hr half life) as the liquid was diluted by fresh condensate.

Particulate-Associated Iodine (Figure 2.3) decreased with a 50 min half life for about one hour, then with a 14 min half life until its concentration reached a value about one third that of elemental iodine after which it decreased slowly at about the same rate as elemental iodine.

This behavior is consistent with previous observations and is explained as follows. The iodine was released to containment environment chiefly in the elemental form, with less than 5% attached to particles. This was substantiated by both Maypack samples taken from the delivery line and the purple color observed in the containment vessel. However, in the saturated steam-air atmosphere, a fog rapidly formed and elemental iodine was adsorbed in the fog droplets, thus forming additional particulate-associated form. Thus, there was a source term for formation of particulate-associated iodine as long as the elemental iodine concentration was high. After 1 hr, the concentrations of the two species were equal and at subsequent times they remained in dynamic equilibrium.

Methyl iodide concentration (Figure 2.4) decreased slowly during the entire test. After a somewhat higher rate initially due to diffusion into other rooms and adsorption by the steam condensate film, it averaged a 30 hr half life. This is considerably faster removal than would be predicted by physical adsorption in water and may be caused by chemical reaction or adsorption on the painted surfaces.

The total iodine concentration as plotted in Figure 2.5 is the sum of all forms of iodine plus the small fraction plated out on the inlet to the Maypacks. Its behavior reflects that

of its constituents. At early times the total iodine concentration decreased about like elemental iodine, since that was the dominant form. At long times it behaved as methyl iodide because it was >90% methyl iodide.

Cesium concentration is plotted in Figure 2.6. It decreased exponentially with a 42 min half life for 2 hr, then more slowly with a 1.9 hr half life until background was reached at 0.1% of its initial concentration. Wall condensate samples showed that the majority of the cesium was transferred from the gas to the vertical walls. Diffusiophoresis could account for a rate equivalent to a 10 hr half life. Therefore, turbulent diffusion must have been the major transfer mechanism.

Uranium concentration was measured by alpha counting the first filter in each Maypack. Determination of the counting efficiency was by calibration against fluorometric analysis. Figure 2.7 shows that the uranium concentration decreased exponentially with a half life of 53 min for about 2 hr, then with a 100 min half life until background was reached.

Distribution within Containment as a Function of Time. Figures 2.8 and 2.9 show the distribution of iodine and cesium, respectively, within the containment complex as a function of time. Both materials were transferred rapidly from the gas phase to structural surfaces and then slowly washed into the vessel sumps. After 24 hr, 90% of the cesium was in the sumps, but only 25% of the iodine. The 75% of the iodine remaining on the walls was not removed by three additional days of steaming and water spraying. Earlier tests⁽¹⁾ had given 35 to 40% of the iodine as the permanently fixed fraction.

Temperature and Velocity Gradients. The velocity due to natural convection was measured by heated thermopile anemometers⁽¹⁾ at various locations. It varied from a maximum

FIGURE 2.8 Distribution of Total Iodine in Containment - Run A-5

FIGURE 2.9 Distribution of Cesium in Containment - Run A-5

upward velocity of 500 ft/min at the vessel centerline to a maximum downward value of 150 ft/min at 1 in. from the side walls.

The temperature in the bulk gas space in the main room was quite uniform, with 7 °F maximum range observed. A sliding thermocouple was used to probe the boundary layer near the wall. A drop of 1.0 °F was measured, half of which occurred at 3/4 in. from the wall.

Fission Product Leakage. Experimental measurement of the amount of fission product materials which accompany air and steam leakage through natural leak points in a containment vessel is difficult because of the very small leak rates involved. For example, during Run A-5, preparations were made to sample a natural leak at one of the thermocouple penetrations. As the vessel was heated by injecting steam, the leak rate at this penetration diminished to a point where it was no longer useful as a sample point.

Information was obtained on leakage through an artificial leak, made by cracking a 1/4 in. valve attached to a 2 in. stub on the penetration panel. The temperature of the leak restriction was intermediate between the containment atmosphere (250 °F) and room temperature. The gases which leaked through the valve were passed through a collection train, including a condenser, Maypack, and rotameter. The leak rate was kept at 1 liter/min of air. Four sequential samples were made by replacing the traps occasionally.

Table 2.2 lists the results. Column 3 is the mass of fission product material collected during the sampling period. Column 4 is the calculated amount expected from the concentration in the bulk gas inside the containment vessel integrated over the sampling period multiplied by the volumetric leak rate. The fourth column lists the decontamination factor (DF)

defined as the ratio of calculated inlet to measured outlet mass. A DF greater than unity therefore indicated that mass was retained in the leak path.

TABLE 2.2. *Fission Product Leakage, Run A-5*

<u>Sample Number</u>	<u>Sampling Period, min after t_0</u>	<u>Micrograms Collected</u>	<u>Micrograms Expected</u>	<u>DF</u>
<u>Total Iodine</u>				
1	23-38	450	745	1.65
2	48-63	211	252	1.19
3	73-103	173	71	0.41
4	168-308	<u>98</u>	<u>25</u>	<u>0.26</u>
Total		932	1093	1.17
<u>Total Cesium</u>				
1	23-38	9.86	10.9	1.10
2	48-63	5.97	7.40	1.24
3	73-103	6.10	9.13	1.50
4	168-308	<u>3.74</u>	<u>1.38</u>	<u>0.37</u>
Total		25.67	28.81	1.12

Table 2.2 shows the interesting, but not surprising, fact that at early times decontamination occurred in the leak path for both total iodine and cesium, while later the leakage became enriched in both materials. However, the overall DF for both materials measured during all four periods shows a slight decontamination effect. The conclusion is that for relatively large leak points (0.3% per day nominal leakage rate) iodine and cesium leak at about the same overall rate as the noncondensable gases.

Conclusions. This experiment showed, as had earlier tests, that aerosols and vapors released into a vessel containing steam and air were mixed very rapidly and that the atmosphere

within any single compartment could be considered homogeneous. Concentrations can differ greatly between connected compartments, however. This information substantiates a basic assumption used in most transport models.

The experimental results for elemental iodine were in excellent agreement with theory based on natural convection and heat-mass transfer analogy. All materials except methyl iodide were effectively removed by natural processes until concentrations were less than 0.1% of their initial value. About 60% of the methyl iodide was removed from the gas phase by some mechanism not presently understood.

Iodine and cesium leakage through a relatively large leak point showed an initial depletion followed by a later enrichment compared to the volumetric rate of noncondensables leaking. The overall decontamination factor for the first five hours was about unity.

CSE Run A-6

The chief objective of Run A-6 was to determine the effectiveness of a containment spray for reducing the airborne concentration of fission product materials, and to compare the results against mathematical models. Test conditions for Run A-6 are listed in Table 2.1. This experiment duplicated Run A-4 made in December, 1967⁽²⁾ in every respect except that the atmosphere was a steam-air mixture instead of room temperature air.

The results of Run A-6 are being evaluated, but the preliminary conclusion is that the spray efficiency in the steam-air atmosphere was about the same as in the earlier tests which used room temperature air. Figure 2.10 shows the concentration of elemental iodine in the gas phase within the main room as a function of time. The 12 min half life during the prespray period agrees well with the 14 min value predicted. The first spray period lasted 10 min and resulted in an effective half

FIGURE 2.10 Elemental Iodine - Run A-6

life of 1.9 min. Allowing for the removal by natural processes during the spray, the half life due to spray operation was 1.92 min. This is to be compared with 1.76 min predicted from absorption theory with gas phase diffusion limiting. This is considered to be a satisfactory verification of the model, since uncertainties in the physical properties required for the model (diffusivity, viscosity, drop size) are greater than the 10% observed difference.

As in the previous experiments with room air⁽²⁾ the second spray period did not maintain this rapid removal. This is attributed to desorption of previously absorbed iodine. The first two spray periods used fresh solution. The third period recirculated the used solution from the sump for a 1 hr period to insure complete equilibrium. The overall decontamination factor for elemental iodine was 2000. This is to be compared with 900 predicted on the basis of the gas-liquid distribution coefficients for hydrolysis only,⁽³⁾ and is considered satisfactory agreement.

CSE Run A-7

The chief objective of Run A-7 was to demonstrate the removal of iodine by a spray solution for which liquid resistance to transfer could be important. The solution used was boric acid in demineralized water. No other additives were used and the pH was 4.8. All other test conditions were the same as in Run A-6. The conditions are listed in Table 2.1.

A secondary objective was to determine the removal of methyl iodide from a steam-air atmosphere by basic sodium thiosulfate. For this purpose a fourth spray period using a sodium thiosulfate solution was added late in the experiment after the chief objective had been accomplished, but while most of the methyl iodide remained airborne.

At the time of writing, the Maypack clusters are being removed, so that the results are not available. However, "thief" samples taken at a single location showed that elemental iodine was removed with a 2.5 min half life during the first boric acid spray and that the total decontamination factor for elemental iodine was about 1000 due to the boric acid sprays. Methyl iodide was not removed. The basic thiosulfate spray removed methyl iodide with a 130 min half life. These results should be considered tentative until a complete evaluation of all sample analyses is made.

Mathematical Models for Fission Product Transport

Natural Transport - R. K. Hilliard and J. G. Knudsen

The natural convection model was described in abbreviated form in the previous progress report.⁽⁴⁾ A report of the complete model is in rough draft form. A comparison of model predictions with experimental results from the seven large-scale tests made to date is given in Table 2.3 for elemental iodine.

TABLE 2.3. *Comparison of Natural Convection Model Predictions with CSE Experiments for Elemental Iodine Concentration Time Dependence*

CSE Run Number	Initial Gas Concentration Half Life, min		Ratio, Predicted to Measured
	<u>Predicted</u>	<u>Measured</u>	
D-1	7.8	8.0	0.98
D-2	7.8	10.0	0.78
A-1	8.3	16.0 ^(a)	0.75 ^(a)
A-2	8.3	10.0	0.83
A-5	14	13.5	1.03
A-6	14	12	1.17
A-7	14	15	<u>0.93</u> ^(b)
			0.92 Avg.

(a) *A source of elemental iodine by desorption from fog droplets not considered in model. Half life for particulate-associated iodine was 11.0 min.*

(b) *Tentative*

Agreement of theory with experiment is good, especially for the latter three experiments in which the containment vessel walls were thermally insulated and consequently for which more accurate heat transfer predictions could be made. Although the model has not been verified completely, it provides a means of predicting iodine behavior in any containment vessel in which engineered safeguards are not operated and a means of understanding its behavior in research experimentation.

Particle Removal by Spray Drops - J. D. McCormack

Significant particle removal by sprays from various accident environments has been observed in CSE Runs A-3, A-4, and A-6. Removal of particles by the falling spray drops is due to three processes: (1) impaction, (2) interception, and (3) diffusion. The importance of each process is dependent upon the size of the particles and the drops; however, the removal rate can be shown to have a minimum value for any drop size. The existence of this minimum permits conservative calculations to be made even in the absence of any particle size information.

The fraction of initial aerosol particles remaining airborne after operation of the sprays, if the gas space to be well mixed and an instantaneous aerosol release is assumed, can be expressed for impaction and interception as

$$\frac{C}{C_0} = \exp - \left[\frac{3 F h E t}{2 V d} \right] , \quad (1)$$

where F = spray volumetric flow rate, ft^3/sec

h = drop fall height, ft

E = sum of single-drop capture coefficients for
impaction and interception

V = gas volume, ft^3

d = spray drop diameter, ft

t = time sprays operated, sec .

E decreases as the particle size decreases. However, the particle diffusion coefficient, D, increases with decreasing particle size according to Equation (2).

$$D = \frac{RT k_m}{3\pi N \mu d_p}, \quad (2)$$

where R = gas constant = 4.97×10^4 lb ft² sec⁻²
lb-mole⁻¹ °R⁻¹

T = temperature, °R

k_m = Cunningham correction factor

N = Avogadro's number = 2.725×10^{26} lb-mole⁻¹

μ = gas viscosity, lb ft⁻¹ sec⁻¹

d_p = particle diameter, ft

The diffusion of the particle to the spray drop through the gas boundary layer surrounding the falling drop can result in a net removal of the aerosol from the gas phase. This removal rate can be expressed in terms of a mass transfer coefficient, and if the sticking coefficient is unity, the Ranz and Marshall correlation can be used to determine this coefficient:

$$\frac{k_c d}{D} = 2 + 0.6 N_{Re}^{1/2} N_{Sc}^{1/3}, \quad (3)$$

where k_c = gas phase mass transfer coef., ft/sec

N_{Re} = Reynolds number = $\frac{d U_t \rho}{\mu}$

U_t = spray drop velocity, ft/sec

ρ = gas density, lb/ft³

N_{Sc} = Schmidt number = $\frac{\mu}{\rho D}$

The fraction remaining airborne, considering only diffusion as the removal mechanism, is

$$\frac{C}{C_o} = \exp \left[- \frac{6 F h k_c t}{d U_t V} \right] \quad (4)$$

Equations (1) and (4) can be combined if the processes are independent:

$$\frac{C}{C_o} = \exp \left[- \frac{3 F h}{2 d V} \left(E + \frac{4 k_c}{U_t} \right) t \right] \quad (5)$$

The single drop efficiency E can be calculated by the method of Ranz and Wong⁽⁵⁾ for particles of various densities. Figure 2.11 shows the effectiveness of the three capture methods for drops falling at terminal velocities. Note the minimum removal at about 0.08 μ particle diameter. For a given spray flow rate, change of drop diameter will affect the removal coefficient as about $1/d^2$ for particles in the interception or diffusion region. For larger particles the removal varies nearly as $1/d$. Note also that the particle density is important only over a limited particle size range of 0.7 to 3 μ diameter with spray drop size of interest. Figure 2.11 was calculated for a 50 gal/min spray falling 33 ft at terminal velocity in the CSE Vessel.

To calculate the particle removal it is obviously necessary to know the drop size as well as the particle size. Drop size information is available from the nozzle manufacturer and in addition we will attempt to confirm the drop size distribution for some selected nozzles. A photographic method, while tedious, seems to have the least chance of error. The particle size is presently measured by use of cascade impactors. However, the accuracy of measurements obtained by sampling from a saturated steam-air atmosphere has not been established.

The uranium and cesium removal due to the spray operation in Run A-4 resulted in concentration half times of 8 to 14 min

FIGURE 2.11. *Calculated Particle Removal in the Main CSE Containment Vessel with a 50 gal/min Spray*

for uranium and half times of 3 to 10 min for cesium. From Figure 2.11, particles of 0.7 and 2.0 μ , would give the observed removal. These sizes are in satisfactory agreement with the measured sizes of 0.6 and 1.5 μ , and indicate that the impaction was the predominant removal mechanism. Data from Run A-6 show similar removal rates in the presence of the condensing steam.

Small-Scale Fission Product Transport Experiments - R. H. Bond

Two runs were made in the Aerosol Development Facility (ADF) during the quarter. In both cases a simulant aerosol, consisting of iodine and cesium with fumes from molten uranium oxide and Zircaloy cladding, was injected into an air-steam atmosphere in the stainless steel ADF containment vessel at thermal equilibrium conditions of 80 °C and atmospheric pressure.

The first run (ADF Run S-96) was a base run with conditions changed appreciably from earlier simulant⁽⁶⁾ runs. The changes included vertical, central steam entry instead of horizontal, side entry, and retention of the condensate as a circulating pool throughout the run rather than periodically draining all condensate. Steam entry was the only source of agitation of the atmosphere. This run duplicated a similar run made a year ago (ADF Run S-86).

The second run (ADF Run S-97) was under conditions essentially identical to the first, but with a noncondensing surface inside the containment vessel. This noncondensing surface consisted of an open-ended, vertical, sheet stainless steel cylinder on the center line of the tank having a total surface area half as great as the containment vessel.

Significant details for the two runs, and the earlier run cited, were as follows:

ADF Run	Initial Gasborne Burden, mg/m ³		Initial Half-Life in Atmosphere, min	
	I	Cs	I (Total)	Cs (Filter + Inlet)
S-86	11 ^(a)	120	10	11
S-96	37	360	17	7.8
S-97	79	210 ^(a)	12.5	10

(a) Based on recovery; others based on calculated input.

Evaluation of the two recent runs is not yet complete. Preliminary results appear to show no appreciable effects on aerosol half-lives because of the presence of noncondensing surface.

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

A simulated reactor vessel with a capacity of 150 ft³ of water at 600 °F and a 2500 psi is being used to study the mass flow rates, transient pressures and forces that may result from a loss-of-coolant accident caused by a piping break in an operating power reactor.

The reactor simulator vessel is mounted on a steel framework and is instrumented to measure the reaction forces, coolant weight, liquid level, and the internal pressures and temperatures, while the heated water is blown out through a simulated pipe break created by a burst rupture disk.

Digital Simulation of Transient Depressurization -

A. S. Neuls and R. T. Allemann

The FLASH2 code⁽⁷⁾ has been applied to three CSE bottom blowdown cases. Test Runs B-10, B-15, and B-13 were used and had nominal break sizes of 2 in., 4 in., and 8 in., respectively. The simulation results indicated that a problem existed in

predicting the proper effective orifice coefficient needed for the two-phase critical-flow model used in the code. Best estimates of effective orifice coefficients and single-phase orifice coefficients at the pipe exit are shown in Table 2.4 for the three cases. A reversed trend in the change of effective orifice coefficient with a change in break size was observed when the effective values were compared with the corresponding single-phase values.

TABLE 2.4. *Orifice Coefficients*

<u>Nominal Break Size, in.</u>	<u>Orifice Diam. Pipe Diam, in.</u>	<u>Single-Phase Orifice Coefficient</u>	<u>Effective FLASH2 Coefficient (Two-phase)</u>
2 (1.689)	0.248	0.61 ^(a)	0.82
4 (3.438)	0.513	0.648 ^(b)	0.69
8 (6.813)	1.00	1.0 ^(c)	0.54

a. *Estimated, see Perry, Chemical Engineers Handbook, 3rd ed., McGraw-Hill, p. 405. 1950.*

b. *Calculated from pressure-time and also from mass flow rate-time data from an ambient temperature blowdown 4 in. diameter*

c. *This neglects the exit pipe entrance losses which are considerable and would give an effective coefficient of 0.61 to 0.70.*

The FLASH2 code used for the simulations is a multinode, digital model of a nuclear power plant coolant loop during the loss-of-coolant accident. The code has enough flexibility to allow its application to the tests of a single leak blowdown for a simple vessel with no internal structure. Some of the important input parameters for the cases are vessel dimensions, initial temperature and pressure, bubble rise velocity, quality gradient factor, and orifice coefficient. The code uses Moody's two-phase critical-flow model of flow through a nozzle⁽⁸⁾ for the leak rate calculations.

The CSE blowdown with different nozzle sizes and initial pressures were calculated using a single node to represent the vessel. The vessel was approximated by a right-circular cylinder 9.35 ft^2 cross section area and 16.25 ft high. Results of the calculations are shown in Figures 2.12, 2.13, and 2.14 for various adjustments of the input parameters.

In the case of Run B-15 (Figure 2.13), a two volume approximation was tried using the same conditions as the single volume case, but with the addition of a 6 ft^3 pressurizer. The pressurizer was initially one-third full and the fluid was at saturated conditions. This case was added in an attempt to simulate the small gas volume that was in the vessel at the start of the experimental run.

The results indicated that the pressurizer volume had little effect, but the quality gradient factor and the increased bubble rise velocity improved the prediction considerably by shifting the elbow in the pressure curve. (The premature cut-off in the third approximation was caused by a calculation instability.)

By comparing, for each blowdown case, the best FLASH2 simulation that used the single-phase orifice coefficient with the experimental results, and referring to the best effective orifice coefficients listed in Table 2.4, it can be seen that a major problem existed in predicting the proper orifice coefficient. The single-phase coefficient increased with orifice size, but the best effective coefficient for the FLASH2 simulations showed an inverse trend. Orifice coefficients are tabulated in hydraulics tests for single-phase flow. The use of a single-phase coefficient in a two-phase situation was, however, questionable.

A possible contributing cause to the problem of defining the proper orifice coefficient was the apparent inability of

FIGURE 2.12 CSE Blowdown Pressure History, Run B-10, $D_B = 1.689$

FIGURE 2.13 CSE Blowdown Pressure History, Run B-15, $D_B = 3.438$

FIGURE 2.14 CSE Blowdown Pressure History, Run B-13, $D_B = 6.803$

FLASH2 to handle a blowdown calculation with a length of pipe extending from the vessel exit. The FLASH2 results presented all use a vessel leak calculation in which losses in the exit pipe were neglected. For the 2 in. break, the physical situation closely matched the FLASH2 model since the mass velocity in the exit pipe was low compared to that at the orifice. Frictional losses in the pipe were, therefore, low. However, for the 8 in. break, the entrance and friction losses were not negligible. Two-volume FLASH2 simulations of the CSE vessel were attempted, using the exit pipe as the second volume, but due to the small relative volume of the pipe, computational instabilities resulting in error terminations were encountered. It should be noted, however, that the single-volume 2 in. break size simulation results using the single-phase coefficients were not close to the experimental results despite the fact the model seemed to fit the physical situation.

The changes in bubble rise velocity and quality gradient factor had a small effect on the results in the ranges considered. For the smaller breaks a bubble rise velocity of 3 ft/sec and a quality gradient factor of 0.8 gave the best agreement to the experimental data when combined with the best coefficient. However, for the 8 in. break a homogeneous steam distribution in the liquid ($C_x = 0$) and a bubble rise velocity of 2 ft/sec gave a closer approximation to the pressure curve shape (Figure 2.14) although the total blowdown time was about the same.

The prediction of the best effective orifice coefficient was, then, the primary problem associated with the use of FLASH2 for describing CSE blowdowns. The results using single-phase orifice coefficients were, however, as accurate as previously reported FLASH2 simulations of simple blowdowns.⁽⁹⁾

It has just been shown the mass flow velocity predicted by the Moody equilibrium-slip-flow model can be adjusted

to give a good fit to the data of the pressure in the vessel, if a nozzle discharge coefficient is used. As also described in previous reports,⁽¹⁰⁾ the mass velocity and thereby the coefficient decreases with increased nozzle size. An empirical expression has been developed for the coefficient for saturated blowdown based on the ratio of outlet (break) area to vessel cross-sectional area.

The expression is,

$$C_{CSEM} = 0.287 R^{-0.165}$$

where: C_{CSEM} = flow coefficient to be applied to the Moody⁽⁸⁾ model of mass velocity based on CSE data.

$$R = \frac{\text{Area of Break}}{\text{Area of Vessel}}$$

This coefficient expression approximates reasonably well the break area effect as measured by other experimenters.⁽¹⁰⁾

Liquid Level During Blowdown

A time-domain reflectometer has been used in the CSE vessel bottom blowdown experiments to measure the liquid level in the vessel during the expulsion of the fluid. Figure 2.15 shows the liquid level fraction as a function of a dimensional-time parameter which is used to correct for outlet area and initial mass differences.

The figure shows that the larger nozzle size has a relatively slower rate of level decrease. This is partly the result of the lower mass velocity (mass flow rate per unit area) of the larger areas which was discussed in the last section.

Moody⁽¹¹⁾ has made some preliminary calculations of the liquid level in a partly full reactor vessel during a bottom break blowdown. Although his results are not directly

FIGURE 2.15 Liquid Level Decrease Versus Dimensional Time

applicable because they were calculated for a vessel initially only 75% full of liquid, they indicate that a steam dome may have a major effect since his predictions show the level dropping nearly linearly with time at first. The CSE blowdowns were done with the vessel essentially full (0.25 vol% nitrogen pressurization dome), and no level surges were observed. Also, the initial decrease was proportional to the square of time. The nitrogen dome used in the CSE experiment did not have significant influence on the liquid level decrease although it prevented the immediate reduction of pressure to saturation that should occur without the gas. Subcooled flow should let the level come down faster than for saturated flow since no bubbles would be formed in the vessel liquid. This trend was not shown by the data and may have been counteracted by a temperature effect. The higher temperature runs have a slightly lower rate of level decrease which may be caused by the exit-choking-flow-rate being less at the higher enthalpy.

To gain insight into the nature of the liquid-vapor action in the vessel, we have replotted some of the level data in Figure 2.16, taking into account the flow coefficient by multiplying by the Moody flow correction coefficient C_{CSEM} . It is seen that liquid level data still differentiates itself into groups depending on the break-to-vessel area ratio. The relatively slower rate of decrease (Figure 2.16) in liquid level for the larger break areas is only partly accounted for by the decreased mass velocity for larger size breaks. This might be explained by the shorter time available for bubble disengagement in the shorter, larger blowdowns; it might be caused by a difference in bubble rise velocity resulting from a larger number of small bubbles being formed in the case of rapid blow-down versus a fewer number of larger, faster rising bubbles being formed during the smaller break blowdown. Since the flow coefficient was taken from comparisons with the FLASH code,

FIGURE 2.16 Corrected Liquid Level Decrease During Bottom, Saturated Blowdown

which assumes a constant bubble rise velocity, the coefficient is probably not independent of this assumption.

Because the level data show differences of up to 100% in the time to reach a certain level (depending on break size) a more detailed theoretical study is being made of the relationships between leak rate, position, and vapor-liquid action in the vessel.

Stress Levels in Vessel Wall - A. J. McElfresh and
V. B. Watwood

The simulator vessel was inspected (radiograph and ultrasonic) after the structural failure of the support system. During this inspection, an ultrasonic defect indication was discovered in the bottom head of the reactor simulator vessel near the outlet nozzle. This defect area became detectable during the course of blowdown experiments since it was not found when the vessel was fabricated and accepted under ASME nuclear vessel standards.

An attempt is being made to determine (analytically and experimentally) the stress levels in the metal which could cause propagation of such an existing defect.

Preliminary analyses of an axial loaded nozzle attached to a sphere are being made. Strain gage values for an axial loaded condition are being used to compare the gage read-out with theory and give an indication of the confidence that can be placed, both on the strain gage readings and on a theoretical treatment. Possible errors in placement or hookup of strain gages and approximations used in the theory suggest that neither method can be used as a basis for calibrating the other, but that they will compliment each other to give a degree-of-correctness indication.

Further results will be used, in part to control the loading of an additional test. This test will consist of applying a bending moment on the nozzle 100 times. The purpose of this test is to see if a pure mechanical loading resulting in near limiting stress levels in the vessel or nozzle will propagate the defect. The loading condition (a moment applied to a nozzle on a hemispherical shell) is complicated and an analysis of this asymmetrical problem is not yet available. The experimental stress values and the predicted stress relationships will be used to assure that the loading applied during the bending test does not exceed the acceptable stress levels. Ultrasonic tests of the defect region will be performed after the loading tests to see if the defect has grown.

PARTICLE TRANSPORT TO SURFACES BY CONDENSING STEAM*

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

Introduction

Following a loss-of-coolant accident in a water cooled reactor, condensation processes can play an important role in removing airborne fission products from the containment volume. Particles and vapors will be transported with saturated steam to the cooler surfaces of walls, droplets, ice (provided in some designs), and reactor components. As the steam condenses, part of the fission gases and the particles will be transferred to these surfaces.

The purpose of the present investigation is to develop a quantitative understanding of the processes causing particle capture in condensing-steam systems. This study is complementary to work being carried out in larger-scale containment demonstration facilities such as LOFT, CSE, and NSPP.

* *This study is reported in this quarterly report for the first time. Some background information is included to better acquaint the reader with the scope and earlier findings of this study.*

In contrast to the larger-scale experiments, this investigation focuses attention on the basic physical processes that produce the aerosol removal. Carefully planned and precisely controlled experiments are needed to permit valid comparison of particle behavior with mathematical models developed in terms of basic transport and thermodynamic properties of the systems.

Information developed should provide a basis for explaining the complex behavior encountered in larger systems, and should provide a reliable procedure for extrapolation and scale-up.

The present investigation embraces three important segments.

1. A detailed review of existing literature on aerosol transport, steam condensation, and related topics.⁽¹²⁾
2. A theoretical treatment of the overall problem.
3. An experimental study of aerosol transport in a condensing steam system.

It is important that 2 and 3 be conducted simultaneously if optimal results are to be attained.

The literature survey indicated that condensation of steam in a containment vessel occurs in a way such that it may be modeled successfully using a boundary layer approach. This is enabled because of the large amount of bulk mixing that characterizes containment vessels during postaccident conditions. Evidently, such systems are further characterized by naturally convected, gaseous boundary layers, which may be laminar or turbulent in nature depending upon physical conditions and vessel geometry.

The literature review emphasized the lack of theoretical and experimental information regarding particle transport in condensing steam. The turbulent flow regime has been given very little attention.

Rigorous Analysis of Laminar Boundary Layers

A thorough investigation of the laminar, naturally convected, condensing-steam boundary layer was conducted. This study was necessary because of the basic information concerning laminar boundary layers, and because of the insights this information could provide regarding turbulent boundary layer behavior. This investigation has its basis in the paper by Minkowycz and Sparrow,⁽¹³⁾ who obtained a general similarity transformation that converted the boundary-layer equations of conservation to

$$\text{(Momentum)} \quad (\pi F')'' + 3F(\pi F')' - 2\pi F'^2 + \left(\frac{1 - \pi}{\pi}\right) = 0 \quad (1)$$

$$\text{(Mass)} \quad \xi'' + 3Sc F \xi' = 0 \quad (2)$$

$$\text{(Energy)} \quad \tau'' + \tau' Pr_{\infty} (3F + \left|\frac{\phi C}{Sc}\right| \Delta W \xi') = 0 \quad (3)$$

Minkowycz and Sparrow obtained specific solutions to the somewhat more sophisticated versions of Equations (1), (2), and (3). However, the physical conditions chosen did not match those of interest in the present study. For this reason, new solutions to Equations (1), (2), and (3) were obtained, which were more closely representative of containment system, postaccident conditions. Calculation was conducted for bulk-interface temperature differences of 1, 2, 4, 8, and 16 °F. Bulk conditions are listed as follows:

1. $P_{air_{\infty}} = 14.696 \text{ psi}$, $P_{steam_{\infty}} = 17.346 \text{ psi}$, $T_{\infty} = 179 \text{ °F}$
 2. $P_{air_{\infty}} = 14.696 \text{ psi}$, $P_{steam_{\infty}} = 14.696 \text{ psi}$, $T_{\infty} = 212 \text{ °F}$
 3. $P_{air_{\infty}} = 14.696 \text{ psi}$, $P_{steam_{\infty}} = 29.305 \text{ psi}$, $T_{\infty} = 249 \text{ °F}$
- (4)

Solution of (1), (2) and (3) was carried out using a Beckman 1137 analog computer. Solutions took the form of velocity, temperature, and concentration profiles across the boundary layer. Representative solutions are given in Figures 2.17, 2.18 and 2.19.

Semi-Empirical Extensions of Boundary Layer Theory

Possession of analog-computer solutions to the boundary-layer problem has enabled a limited investigation to determine the validity of extending information concerning simple heat transfer to situations wherein condensation occurs. Advantage is taken, in extending these equations, of the physical and mathematical similarity between the processes of heat conduction and molecular diffusion.

The differential equations describing simple heat transfer and transfer through condensation are almost identical when the dependent variables in them are appropriately nondimensionalized. In order to obtain the proper fluxes at the boundaries, however, a correction must be made for the nonsimilar boundary conditions. In the pure heat transfer case the velocity normal to the vertical surface is zero, but a nonzero normal velocity must accompany mass transfer.

The resulting equation for the mass flux to the surface is,

$$\dot{m} = \left(\frac{p_1}{p_2} + 1 \right)_i \left(\frac{D_{12}}{T} \right)_f \frac{M_1}{R} \frac{N_{Sh}}{H} \frac{\phi}{e^{\phi} - 1} [(p_1)_{\infty} - (p_1)_i] \quad (5)$$

where p_1 = vapor pressure of steam
 p_2 = vapor pressure of air
 D_{12} = mutual diffusive coefficient of steam and air
 T = absolute temperature
 M_1 = gram-molecular weight of steam
 R = universal gas constant
 H = height of vertical surface

2.43

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Neg 0684062-3

FIGURE 2.17 Computed Velocity Profiles Adjacent to a Flat, Vertical Plate

Neg 0684062-2

FIGURE 2.18 *Computed Steam Concentration Profiles Adjacent to a Flat, Vertical Plate*

Neg 0684062-1

FIGURE 2.19 Computed Temperature Profiles Adjacent to a Flat, Vertical Plate

- i = property at liquid-vapor interface
- f = property at mean boundary layer temperature
- ∞ = property in bulk exterior of boundary layer
- N_{Sh} = Sherwood number or dimensionless mass transfer
- ϕ = profile correction factor for mass transfer.⁽¹⁴⁾

The Sherwood number is found from the analogy with heat transfer,

$$N_{Sh}(N_{Gr}, N_{Sch}) = N_{Nu}(N_{Gr}, N_{Pr}) \quad , \quad (6)$$

i.e., the functional form of the Sherwood number is identical to that of the Nusselt number, or dimensionless heat transfer, with the Schmidt number replacing the Prandtl number and the appropriate Grashof number being used.

Calculations were made using both an empirical,⁽¹⁵⁾

$$N_{Nu} = 0.59 [N_{Gr} N_{Pr}]_f^{1/4} \quad , \quad (7)$$

and an analytical relation,⁽¹⁶⁾

$$N_{Nu} = 0.517 [N_{Gr} N_{Pr}]_f^{1/4} \quad (8)$$

for the dimensionless heat transfer. Since the analytical relation was calculated by a boundary layer method similar to that used for the analog computer solution, it is more appropriate for comparison. For the cases investigated in this study a discrepancy of less than 3% was found between the analog computer results and those utilizing Equation (8). No definitive experimental studies are known which can be used for comparison.

Extension of the Rigorous Boundary Layer Theory to Account for Aerosol Behavior

A similarity transformation of the equation expressing the conservation of aerosol particles was used in deriving an ordinary differential equation which may be solved in conjunction with Equations (1), (2) and (3) to provide a description of aerosol deposition behavior. In its most general form, this equation may be written,

$$(\pi T') = \phi_1 \cdot (\pi T') + \phi_2 \cdot \left(T + \frac{r_i}{r_\infty - r_i} \right) \quad (9)$$

where

$$r = P/\pi$$

P = mass fraction of particles,

$$T = \frac{r - r_i}{r_\infty - r_i},$$

$$\phi_1 = \phi_1(F, \phi, \xi, \xi'), \text{ and}$$

$$\phi_2 = \phi_2(F, \phi, \pi', \pi, \xi, \xi', \xi'').$$

Major assumptions used in the formulation of Equation (9) are:

1. The aerosol moves in the free-molecule regime, where bulk motion in the y -direction is described by $\bar{v} + \bar{v}'$, where $\bar{v}$ is the molar-average velocity, and $\bar{v}'$ is the diffusiphoretic correction factor given by Waldman.⁽¹⁷⁾
2. Aerosol motion in the x -direction is equal to the mass-average velocity, of fluid in that direction.
3. Particle diffusion coefficients are given by the modified Stokes-Finstein relationship.
4. The mass-fraction of particles is very small.

The algebraic nature of ϕ_1 and ϕ_2 is complex, and it is therefore expedient to utilize a digital method for their evaluation. A hybrid-computer routine is currently being prepared to solve Equation (9). In this routine, the original boundary-layer analog supplies $F, \phi, \pi, \pi', \xi, \xi'$,

and ξ'' to the digital segment of the computer. This information is used to compute ϕ_1 and ϕ_2 , which, in turn are transferred across the interface to the analog segment. The analog computer then solves (9), providing a computed profile of the aerosol concentration.

Experimental Design

An experiment was designed to measure heat, steam, and aerosol transport across a naturally-convected boundary layer. A schematic of the arrangement is shown in Figure 2.20. A condensation plate is exposed to a steam-air atmosphere maintained within an insulated pressure vessel. Conditions of pressure, temperature, and humidity are set and held constant using appropriate sensors and controls. Aerosol is generated and introduced to the chamber atmosphere via a recycle stream.

The condensation plate consists of ten segments, mounted on a vertical rack as a total unit. A constant temperature is maintained across the plate surface by circulating coolant in a serpentine pattern individually through each segment. Heat loads to each plate would be determined by measuring entrance and exit temperatures and flow rates. A sensitive thermistor-type thermometer system has been obtained for this purpose.

Temperatures at the surface of the plate, in the bulk, and across the boundary layer are to be measured with thermistor-type sensing elements. Velocity and turbulence of the boundary layer would be measured with a hot-wire anemometer, which has been obtained for this purpose.

Segments of the plate fit together with a semi dovetail configuration. Any two segments of the plate can be displaced slightly, thereby creating a gap which allows collection of liquid run-off from all higher plates. This liquid plus any

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FIGURE 2.20 Schematic of Apparatus

BNWL-926

entrained aerosol is channeled to the rear of the collecting plate and withdrawn for analysis.

Dimensions of the system were specified on the basis of desired objectives and cost considerations. Width of the plate (3 ft) was set large enough to ensure that edge effects would be negligible. Length of the plate (10 ft) was specified to guarantee its ability to generate a turbulent boundary layer. Conditions for establishment of turbulence were determined using standard methods. Figure 2.21 indicates conditions for which turbulent regime is expected to exist, and shows that a plate length of at least ten feet is necessary to ensure good measurement.

This experiment appears to be unique, in that it allows, by virtue of its segmented plate, the differential analysis of heat, steam, and aerosol transport. It is unique also because its dimensions permit analyses of turbulent boundary layers in condensing-steam systems -- an area that is virtually devoid of information at the present time.

This experiment was entering the final stage of its design when curtailment of funds forced its indefinite suspension.

References

1. Nuclear Safety Quarterly Report for July through October 1967, BNWL-754. Pacific Northwest Laboratory, Richland, Washington, May 1968.
2. Nuclear Safety Quarterly Report for November, December 1967, January 1968, BNWL-816, p. 2.1. Pacific Northwest Laboratory, Richland, Washington, September 1968.
3. A. E. J. Eggleton, A Theoretical Examination of Iodine-Water Partition Coefficients, AERE-R-4887. United Kingdom Atomic Energy Authority, Atomic Energy Research Establishment, Harwell, Berks, England, February 1967.
4. Nuclear Safety Quarterly Report for May through July 1968, BNWL-894. Pacific Northwest Laboratory, Richland, Washington, October 1968.

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FIGURE 2.21 Expected Points of Transition from Laminar to Turbulent Boundary Layer

5. W. E. Ranz and J. B. Wong. Ind. Engr. Chem., vol. 44, pp. 1371-1381. June 1952.
6. R. K. Hilliard, L. F. Coleman, and J. D. McCormack. Comparison of the Containment Behavior of a Simulant with Fission Products Release from Irradiated UO₂, BNWL-581. Pacific Northwest Laboratory, Richland, Washington.
7. J. A. Redfield, J. H. Murphy, and V. D. Davis. FLASH2: A Fortran IV Program for the Digital Simulation of a Multinode Reactor Plant During Loss-of-Coolant, WAPD-TM-666. Bettis Atomic Power Laboratory, Pittsburgh, Pa., April 1967.
8. F. J. Moody. "Maximum Flow Rate of a Single Component, Two-Phase Mixture," Am. Soc. Mech. Engrs., Paper 64-HT-35. 1964.
9. J. A. Redfield and J. H. Murphy. "Comparison of the FLASH2 Loss-of-Coolant Analysis Method with LOFT Semiscale Test Results," Trans. Am. Nucl. Soc., vol. 10, p. 701. November 1967.
10. Nuclear Safety Quarterly Report for July, August, September, and October, 1967, BNWL-754. Pacific Northwest Laboratory, Richland, Washington, June 1968.
11. F. J. Moody. Liquid/Vapor Action in a Vessel During Blowdown, APED-5177. General Electric Company, Atomic Power Equipment Dept., San Jose, California, June 1966.
12. T. W. Horst. A Review of Particle Transport in a Condensing-Steam Environment, BNWL-848. Pacific Northwest Laboratory, Richland, Washington, June 24, 1968.
13. W. J. Minkowycz and E. M. Sparrow. "Condensation Heat Transfer in the Presence of Noncondensables, Interfacial Resistance, Superheating, Variable Properties, and Diffusion," Intern. J. Heat Mass Transfer, vol. 9, pp. 1125-1144. 1966.
14. R. B. Bird, W. E. Stewart, and E. N. Lightfoot. Transport Phenomena, John Wiley and Sons, New York, N.Y. 1960.
15. W. H. McAdams. Heat Transmission, McGraw-Hill, New York, N.Y., 1954.
16. H. Schlichting. Boundary Layer Theory, McGraw-Hill, New York, N.Y., 1962.
17. L. Waldmann. "Uber die Kraft eines Inhomogenen Gases auf Klein Suspensierte Kugeln," Z. Naturforsch., vol. 14a, pp. 589-599. 1959.

3. PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

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

Introduction

Practical methods of further assuring the integrity of nuclear reactor pressure boundaries are of continuing interest from the standpoint of nuclear safety. A nondestructive technique with considerable potential in this area involves detection of "acoustic emission" produced by metal as it deforms and fractures. The technique is applicable on a continuous surveillance basis to detect the formation or growth of flaws and the resolution potential exceeds that of any of the conventional nondestructive testing methods such as ultrasonics or radiography.

Work to develop application of acoustic emission techniques to reactor system integrity surveillance has been sponsored at the Pacific Northwest Laboratory by the USAEC, Division of Reactor Development and Technology since February, 1966. This is related to work sponsored at Phillips Petroleum Company, Idaho Test Site on plant application of developed techniques.

Discussion

Discussion of program progress is presented under the same topical outline used in previous quarterly reports.

Task 1 - Acoustic Emission Signal Analysis and Characterization

It has become evident that some basic work to improve detection methods is necessary to attain the resolution and reliability needed for routine application of acoustic

emission. Techniques currently in general use involving surface mounted accelerometers or ultrasonic transducers are adequate for a relatively noise free condition such as hydrostatic testing and for conditions where only a gross indication of acoustic emission is required. For those conditions, however, where a significant noise field such as that from high velocity fluid flow or cavitation is present and where accurate resolution of the number of emission signals is required, an improved sensing technique is needed. The conventional sensors in general respond at a frequency below 1 MHz which may incur problems from background noise and the response to a given emission signal is so long in time that many closely following signals may be lost in the response to the first signal.

In keeping with the indicated need for improving sensing methods, a study has been in progress to investigate wave propagation from an acoustic emission source (both simulated and real) with the object of better utilization of available sensing methods for detection of acoustic emission. Complete results of this study will be reported in a separate topical report.

Mathematical relationships were developed to define the number of reflected signals from a single source event that would be observed along with the directly transmitted signal in terms of plate thickness, distance from the source, propagation velocity in the materials and couplant, sensor diameter, and frequency. This provided a tool for predicting the length of signal detected with a given sensor from a single source event. The practically unattainable ultimate is to observe only the directly transmitted signal. Briefly, the mathematical relationships show that increasing the sensor size and raising the detection frequency both serve to reduce the angular aperture of the sensor. This in turn reduces the number of signal reflections observed and shortens the received signal.

Fortunately, these requirements are compatible with the practical aspects of emission detection in the field. Reactor noise measurements reported previously⁽¹⁾ and a test of emission detection in the presence of hydraulic noise discussed under "Task 6" of this report both substantiate the need to work in the low megahertz frequency range. A brief study of the acoustic emission signal per se shows that it is not the limiting factor on high frequency detection. Sensors up to 30 MHz were used to study emission, and the signal transmitted in the metal (steel) was sufficiently short to cause the sensor to respond at its fundamental frequency of 30 MHz. This is well above any practical level for field surveillance application because of frequency dependent attenuation in the material.

The different modes of signal transmission (longitudinal, shear, and Rayleigh) were studied for merits of one over the other as a vehicle for emission detection. It appeared that detection of shear waves would involve the least interference from other modes and produce the sharpest signal.

Tests were conducted to confirm the validity of the above conclusions using both an artificial signal source and real acoustic emission. A 1/2 in. thick A-212-B steel plate having an area about 2 ft by 3 ft was used. In one end of the plate a "V" groove was cut across the 2 ft dimension about 6 in. from the end. This 6 in. section was then cut with a band saw to produce a series of 6 in. long by 1/2 in. square cantilever projections. These were bent to produce acoustic emission from deformation and cracking at the root of the projection.

An artificial source was first used in these tests. A small ultrasonic shear wave transducer was mounted on the end of the plate opposite to where the strips had been cut. This

transducer was then pulsed with a short repetitive burst signal which transmitted shear waves into the plate.

As mentioned earlier, it is desirable to use as high a frequency as possible in detecting acoustic emission to produce short, sharp signals for good counting and triangulation. Most industrial steels and stainless steels have high shear wave attenuation due to grain scattering at frequencies of 5 MHz and above. Since it is also desirable to be able to detect emission signals as far from the source as possible, a compromise is necessary. At the present time, no specific attenuation data is available for A-212-B, but it was reasonably assumed that for frequencies below 5 MHz, attenuation would be quite low. The major loss mechanism then becomes the inverse distance loss by the spreading out of waves from a point source.

Frequencies of 2 MHz and 4.5 MHz were chosen for studies since this represents an overall desirable range and appropriate transducers were available. The expected number of reflections and signal durations were calculated for these frequencies, assuming transducers located at 5, 15, and 30 in. distances from the source. Water was used to couple energy out of the plate into the sensors. For water $V' = 0.587 \times 10^5$ in./sec and for A-212-B steel $V = 1.28 \times 10^5$ in./sec. The transducer diameters available for a frequency of 2 MHz were 0.215 and 0.75 in. and for 4.5 MHz, 0.1875 and 0.75 in. Table 3.1 shows the number of reflections $n - 1$ and the expected signal durations Δt , with n and Δt determined by the mathematical relations. As an aid to determining the total signal duration (Δt) from the total number of reflections ($n - 1$), a graph was prepared (Figure 3.1) for the three detection distances selected. This can also be used to determine the time between successive reflections.

The measured pulse lengths generated using the artificial source of shear waves are shown in Figures 3.2 and 3.3. These

can be compared with the calculated values in Table 3.1. The calculated Δt values are seen to compare well with the measured signals.

TABLE 3.1 *Signal Reflections and Durations*

$$V = 1.28 \times 10^5 \text{ in./sec}$$

$$V' = 0.587 \times 10^5 \text{ in./sec}$$

$$\tau = 0.5 \text{ in.}$$

<u>f, MHz</u>	<u>D, in.</u>	<u>X, in.</u>	<u>n - 1</u>	<u>Δt, μsec</u>
2	0.215	5	8.4	14.4
2	0.215	15	27.3	44.5
2	0.215	30	55.6	88
2	0.75	5	3.3	3.5
2	0.75	15	11.9	10.5
2	0.75	30	24.7	21
4.5	0.1875	5	4.8	6
4.5	0.1875	15	16.5	18.5
4.5	0.1875	30	33.9	36.5
4.5	0.75	5	1.9	1.6
4.5	0.75	15	7.6	4.8
4.5	0.75	30	16.1	9.4

$$V = V' = 1.28 \times 10^5 \text{ in./sec}$$

2	0.1875	30	26.4	23.5
4.5	0.1875	30	10.3	4

An important aspect of the measure signals is the fact that the shear waves were mode converted to longitudinal waves and received by longitudinal receivers through a water couplant between the sample and sensor. The transmission factor between a solid (metal) and a liquid (water) for shear waves converting to longitudinal waves varies as a function

FIGURE 3.1. Number of Reflections Versus Time Duration

D = 0.251 inches, X = 5 inches
Calc. $\Delta t = 14.4 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.75 inches, X = 5 inches
Calc. $\Delta t = 3.5 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.215 inches, X = 15 inches
Calc. $\Delta t = 44.5 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.75 inches, X = 15 inches
Calc. $\Delta t = 10.5 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.215 inches, X = 30 inches
Calc. $\Delta t = 88 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.75 inches, X = 30 inches
Calc. $\Delta t = 21 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

FIGURE 3.2. Measured Signals for Artificial Source
at Frequency of 2 MHz

D = 0.1875 inches, X = 5 inches
Calc. $\Delta t = 6 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.75 inches, X = 5 inches
Calc. $\Delta t = 1.6 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.1875 inches, X = 15 inches
Calc. $\Delta t = 18.5 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.75 inches, X = 15 inches
Calc. $\Delta t = 4.8 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.1875 inches, X = 30 inches
Calc. $\Delta t = 36.5 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

D = 0.75 inches, X = 30 inches
Calc. $\Delta t = 9.4 \mu\text{sec}$
Time Scale = $10 \mu\text{sec/cm}$

FIGURE 3.3. *Measured Signals for Artificial Source
at Frequency of 4.5 MHz*

of shear wave incident angle. A small bundle of shear wave rays propagating down the plate approximately parallel to its surfaces without appreciable reflection at the surfaces has a low transmission factor. The transmission factor is higher for a similar bundle propagating at a slightly oblique angle with respect to the surfaces. The first arrival (unreflected signals) are therefore of smaller amplitude than subsequent signals which are from obliquely traveling rays which reflect at the surfaces. The signal thus starts at a low amplitude and builds up with time until it finally decays off due to the increasing refraction angle of subsequent reflections. These rise and decay effects are illustrated in Figures 3.2 and 3.3.

In Figures 3.4 and 3.5 measured signals for actual acoustic emission sources are shown. The sensors were positioned at distances from the source corresponding to those in Figures 3.2 and 3.3. A separate shear wave critical angle sensor was placed as near as possible to the notch to supply a signal for triggering the display oscilloscope. Because each acoustic emission event is expected to be unique, signals at various distances had to be compared by observing the same signal at two locations simultaneously. This was accomplished with a dual beam oscilloscope, and the traces shown in Figures 3.4 and 3.5 were adjusted with independent time delays so the signals appeared together.

Comparing the signals in Figures 3.4 and 3.5 with those of Figures 3.2 and 3.3 shows that the actual acoustic emission signals appear to also be approximately represented by the derived mathematical expressions. It is to be expected, however, that differences should exist between signals from artificial as opposed to real emission sources. This is because an emission burst can occur at any time or position

$\left\{ \begin{array}{l} D = 0.215 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 14.4 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.215 \text{ inches, } X = 15 \text{ inches} \\ \text{Calc. } \Delta t = 44.5 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.215 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 14.4 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.215 \text{ inches, } X = 30 \text{ inches} \\ \text{Calc. } \Delta t = 88 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.75 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 3.5 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.215 \text{ inches, } X = 15 \text{ inches} \\ \text{Calc. } \Delta t = 44.5 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.215 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 14.4 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.75 \text{ inches, } X = 15 \text{ inches} \\ \text{Calc. } \Delta t = 10.5 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

FIGURE 3.4. *Measured Signals for Actual Emission Source at Frequency of 2 MHz*

$\left\{ \begin{array}{l} D = 0.1875 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 6 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.1875 \text{ inches, } X = 15 \text{ inches} \\ \text{Calc. } \Delta t = 18.5 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.1875 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 6 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.1875 \text{ inches, } X = 30 \text{ inches} \\ \text{Calc. } \Delta t = 36.5 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.75 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 1.6 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.1875 \text{ inches, } X = 15 \text{ inches} \\ \text{Calc. } \Delta t = 18.5 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.1875 \text{ inches, } X = 5 \text{ inches} \\ \text{Calc. } \Delta t = 6 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

$\left\{ \begin{array}{l} D = 0.75 \text{ inches, } X = 15 \text{ inches} \\ \text{Calc. } \Delta t = 4.8 \text{ } \mu\text{sec} \\ \text{Time Scale} = 10 \text{ } \mu\text{sec/cm} \end{array} \right.$

FIGURE 3.5. *Measured Signals for Actual Emission Source at Frequency of 4.5 MHz*

(near the notch) whereas the artificial source has periodicity and a fixed position.

In some instances, actual emission signals were barely evident at 30 in. sensor positions. This is not surprising since the acoustic emissions are essentially from point sources. The major loss in energy then is due to the spreading out of the spherical waves. (Since the sensors are pressure sensitive, the loss of signal amplitude varies inversely with distance from the source. For instance, a sensor placed at 30 in. would have one hundredth the amplitude measured at 0.3 in.) Additional losses occur due to material attenuation, and to amplitude loss in the first arrival wave fronts because of transmission factor variations mentioned previously.

A notable improvement in sensitivity was achieved by using direct shear wave sensors in place of the critical angle sensors. About an order of magnitude increase in signal was observed by mounting the shear sensors on the edge of the 1/2 in. plate. The signal also decreased in time duration because the angular aperture is smaller. An example of the signal for an edge mounted 2 MHz shear sensor having about 1/2 in. dimensions is shown in Figure 3.6. In Table 3.1, Δt was also calculated for $X = 30$ in. and $V = V'$, and the result is compared to the observed signal in Figure 3.6. The corresponding signal at $X = 30$ in. for about the same size ($D = 0.215$ in.) critical angle sensor is also shown. (The direct shear sensor is more sensitive. The vertical gains on the two channels of the oscilloscope were adjusted to produce signals of similar amplitude for better comparison of signal length.)

The question arises as to how direct shear sensors can be mounted to attain the additional sensitivity. Since the sensors can be quite small, a shallow hole or indentation which permits the sensor to be recessed may be permissible. Also, the

Edge Mounted Shear Sensor
D = 0.1875 inches, X = 30 inches
Calc. $\Delta t = 23.5 \mu\text{sec}$
Time Scale = 10 $\mu\text{sec/cm}$

Critical Angle Sensor
D = 0.215 inches, X = 30 inches
Calc. $\Delta t = 88 \mu\text{sec}$
Time Scale = 10 $\mu\text{sec/cm}$

FIGURE 3.6. *Edge Mounted Shear Versus Critical Angle Sensor*

geometry of the test sample may provide natural corners for attaching shear sensors in desirable orientations. Other ways of adapting the shear sensor are also being sought.

Transmission and reflection factor studies and other studies involving the efficiency of certain sensor coupling methods are being undertaken. The effects of sample curvature are also being studied.

Preliminary study of acoustic emission from Type 304L Stainless Steel specimens that failed in tension-tension high cycle fatigue showed significant acoustic emission activity well before formation of a visible surface crack. The specimen configuration is shown in Figure 3.7A. This specimen was pre-loaded as shown in the loading diagram (Figure 3.7B) and then cycled at 1800 cycles per minute until failure. Representative acoustic emission produced by these samples is shown in Figure 3.8. Practically no emission was observed until after approximately 6000 cycles. From this point, the emission rate increased gradually until about 22,000 cycles was reached, after which the emission rate increased sharply. A visual surface crack was detected at approximately 34,000 cycles and it grew to 1/8 in. in length by 55,000 cycles. The large fluctuations in the acoustic emission rate after 34,000 cycles are apparently due to the crack growing in steps. It is significant that acoustic emission activity was first evident at 20% of the cycles necessary to form a visible surface crack. Also, the most intense activity occurred in the region from shortly before to shortly after initial formation of a visible crack. Further study is necessary to answer questions such as, "What is the material condition that gives rise to the initial emission activity?", and "Can the emission rate fluctuations be identified with step increments in the fatigue crack?".

A. FATIGUE SPECIMEN

B. SPECIMEN LOADING DIAGRAM

FIGURE 3.7 *Cantilever Beam Specimen Fatigued by Cyclic Loading*

FIGURE 3.8. A Plot of Acoustic Emission from a Specimen Undergoing Fatigue

Task 2 - High Temperature Transducer Development

Transducers used for continuous monitoring of reactor pressure systems for acoustic emission will be required to withstand a combined environment of high temperature and nuclear radiation for periods of at least 1 1/2 to 2 years. No data are available on the ability of lithium niobate piezoelectric material to withstand such an environment. Since this is the primary candidate material for fabrication of high temperature transducers, the need exists for environmental testing. Design of such a test utilizing a test reactor at Washington State University has been completed. With a maximum fast flux density of about 1.4×10^{16} neutrons/cm²/sec pulse operation and about 1×10^{13} neutrons/cm²/sec steady state, the required total exposure to a distribution of thermal and fast neutron can be acquired in less than a days time to give a gross evaluation of radiation effects. The test capsule will be heated during exposure.

Lithium niobate used in the edge mounted shear sensitive mode has been tested for detection sensitivity in the laboratory. Used in this manner, the sensitivity appears comparable to that of PZT-5.

Task 3 - System Analysis Study

No further work has been done on this task this quarter.

Task 4 - Monitor System Development

A solid state emission count summation circuit has been designed and fabricated. This work in companion with the emission rate analyzer to provide a continuous record of both count rate and count summation. In many instances, count summation is a more useful parameter for determining trends than is count rate.

Task 5 - Cooperative Work with Phillips Petroleum Company

Plans for a joint Phillips Petroleum Company-PNL study of background noise on the San Onofre, pressurized water power reactor located south of Los Angeles, California, were temporarily cancelled in late August. The reactor scheduling did not permit sufficient time to install sensors on the system.

Task 6 - Application Engineering

A significant question to be answered on application of acoustic emission is, "Can acoustic emission signals be segregated from the general system noise?" In the case of a nuclear reactor pressure system, the primary concern is with hydraulic noise which might mask acoustic emission. Noise measurements made during May and June 1968 on the Commonwealth Edison Co.'s Dresden I nuclear power reactor indicated that the hydraulic noise would not be a problem if acoustic emission could be detected in the frequency region of about 750 kHz to MHz.⁽¹⁾ The existence of acoustic emission signal components in this frequency range was confirmed in the laboratory. A logical next step was to combine acoustic emission and hydraulic noise to demonstrate the feasibility of segregating acoustic emission signals from the hydraulic noise.

In order to introduce known acoustic emission into a simulated reactor coolant system noise environment, a test specimen was attached to an existing hydraulic test loop. The loop at the point where the test specimen was attached consists of a 2 5/8 in. OD steel tubing. For the purpose of this test, flow velocities up to slightly over 30 ft/sec and fluid temperature of about 500 °F were used. A gate valve located about 10 ft upstream of the point where the specimen was attached was used to generate cavitation. The test specimen consisted of 3/8 in. thick plate of A-212-B carbon steel 40 in. long by 12 in. wide. Figure 3.9 shows the test arrangement. Eight-inch

FIGURE 3.9 *Test Arrangement for Detection of Acoustic Emission in the Presence of Hydraulic Noise*

long "fingers", 1/2 and 3/4 in. wide were cut in one end of the plate for use in generating acoustic emission. These were manually bent down and ultimately broken to produce emission signals. They are shown in the foreground of the right hand picture in Figure 3.9. All surface oxide and scale was removed from the plate in the vicinity of the root of the fingers to assure that true acoustic emission was produced. Sensors were located at the opposite end of the plate with the attachment to the hydraulic loop made between the sensors and emission source at a point 8 to 10 in. from the sensors. The plate was attached to the loop piping by means of two "U" bolts around the pipe and through the plate. General Electric Co. RTV-116 silicone rubber was used for acoustic coupling between the plate and pipe. Previous tests have shown this to be an efficient coupling agent. Also, a groove was cut in the plate to match the pipe radius to increase the contact surface and enhance acoustic transmission.

The test system is outlined in Figure 3.10. The output from the data sensors located at the opposite end of the plate from the bend tabs was fed, two channels at a time, through conditioning electronics and into a dual beam oscilloscope. A single sensor was located very near the root of the bend tabs and its output was fed into the external trigger circuit of the oscilloscope. With this arrangement and the oscilloscope operated in the single sweep, external trigger mode, an acoustic emission signal would trigger the oscilloscope as it passed the first sensor and the complete response of the data sensors to this signal then appears on the screen at a position determined by sweep rate. The data images on the screen were recorded photographically. Data signals viewed in the above manner can be positively identified as being acoustic emission originating at the root of the bend tabs.

3.21

FIGURE 3.10 *Sketch of Test System for Detection of Acoustic Emission in the Presence of Hydraulic Noise*

BNWL-926

The noise spectra produced on the pipe and on the test specimen by turbulent flow of 475 °F water in the pipe at approximately 30 ft/sec velocity are shown in Figure 3.11. As the upper photos show, the major noise field lies below 200 kHz. Since the analyzer (Tektronix Type 1L5 Spectrum Analyzer) is prone to become unstable when overloaded on the input, the noise data were analyzed a second time with a 300 kHz high-pass filter in the circuit. These are shown in the bottom photos and agree well with the broader band analyses. Gain between the transducers and the analyzer was limited to 10^3 for spectral measurements in order to minimize analyzer overload. Comparison of the spectra from the pipe and the test plate show that acoustic coupling was quite good. In fact, the characteristics of the plate appeared to amplify the hydraulic noise, particularly in the lower frequencies.

Introduction of cavitation produced a nominal factor of 50 increase in noise intensity compared to turbulent flow alone. This is illustrated in Figure 3.12 which shows a video trace of the noise signals produced in the test specimen for both conditions. These traces were taken directly from the transducer with no amplifiers or filters between it and the oscilloscope. The greatest increase in noise level produced by cavitation was in the frequency region below 200 to 300 kHz. Spectra comparisons are given in Figure 3.13. With the onset of cavitation, the increase in low frequency noise was so severe that the signal had to be filtered at 500 kHz high-pass with a 24 dB/octave filter in order to prevent instability in the analyzer. Although the major increase in noise produced by cavitation was in the lower frequencies, it also generated a significant increase in the spectrum level to about 800 MHz.

Information on the spectral content of hydraulic noise observed at the Dresden I nuclear power reactor ⁽¹⁾ is reproduced in Figure 3.14 for comparison purposes. In general,

FIGURE 3.11 Hydraulic Noise Spectra Turbulent Flow

Turbulent Flow - No Cavitation

Vertical: 0.005 Volts/cm
Horizontal: 0.001 Sec/cm

Turbulent Flow - With Cavitation

Vertical: 0.2 Volts/cm
Horizontal: 0.001 Sec/cm

1.0 MHz. Longitudinal Transducer, Surface Mounted on the Specimen. No Filter and no Amplification Between Sensor and Scope.

FIGURE 3.12 Comparison of Noise Signal for Cavitation and Noncavitation Conditions

FIGURE 3.13. Hydraulic Noise Spectra Turbulent Flow and Cavitation Conditions

25 Ft/Sec. Flow Velocity
 500 °F Water
 Turbulent Flow
 22" West Recirculation Header

6" By-Pass, "D" Secondary Steam Generator
 High Flow-Main Valve Throttled
 Probable Cavitation
 400 °F Water

FIGURE 3.14 *Hydraulic Noise Spectra Dresden I Nuclear Power Reactor*

the spectral distribution is very similar to that produced in the test arrangement. The spectrum shown in the bottom photo can only be assumed to originate from cavitation. However, it compares quite closely to that from known cavitation shown in the lower left photo in Figure 3.13. Some differences in absolute level between the on-reactor and the test conditions are attributable to differences in lead length between the transducers and preamplifier and overall lead lengths between transducer and analyzer.

Both on-reactor data and test loop indicate that hydraulic noise including that generated by cavitation is primarily below 1 MHz in spectral content. Figures 3.15 and 3.16 present the results of efforts to detect known acoustic emission at a distance of 32 in. in the hydraulic noise environment described above with four different detection systems. Acoustic emission was produced by slowly bending one of the "fingers" on the test plate until an emission event was generated to trigger the oscilloscope and record the detection response on film.

The same detection arrangement was used for the top trace in all photographs in Figures 3.15 and 3.16 to provide a correlation reference. This consisted of a 2.2 MHz shear transducer mounted on the edge of the plate with the sensor output routed through a 1.7 to 2.5 MHz narrow band amplifier which provided a gain of 1.5×10^4 . The oscilloscope sweep rate used was 100 μ seconds/cm. With a distance of about 31 in. between the trigger sensor and data sensors, the time delay for shear signal propagation should be:

$$\frac{31 \text{ inches} \times 10^6 (\mu\text{in./in.})}{13 \times 10^4 \text{ in./sec (shear velocity in steel)}} = 238 \mu\text{sec}$$

The data traces agree very closely with this value.

FIGURE 3.15 *Acoustic Emission Signals Detected in the Presence of Hydraulic Noise-A*

FIGURE 3.16 *Acoustic Emission Signals Detected in the Presence of Hydraulic Noise-B*

In the bottom trace of the two top photographs of Figure 3.15 is shown response of a 1.7 MHz shear sensor mounted on the edge of the plate with the signal conditioned by a broadband amplifier (30 kHz to 3 MHz) with 10^4 gain, and a 1.2 to 2.0 MHz band pass filter. The bottom trace in the bottom photographs of Figure 3.15 shows the response of an identical system except that the sensor is mounted on the edge of a steel block cemented to the surface of the plate.

The bottom trace in all photographs in Figure 3.16 shows emission signals as detected by a 1 MHz longitudinal sensor with the crystal damped and mounted on the plate surface. The sensor output is fed into a broadband amplifier (30 kHz to 3 MHz) for a gain of 10^4 and filtered 600 kHz to 1.2 MHz in one case and 900 kHz to 1.2 MHz in the other.

In summary, the significant features of the acoustic emission detection data are:

1. Detection of acoustic emission above approximately 1.5 MHz appears to be unaffected by hydraulic noise, even cavitation. Detection in this range is not felt to be a limitation. Recent investigations of the emission signal per se indicated that all of the emission at least from steel are sharp rise transients that will produce response in transducers in excess of 30 MHz.
2. The edge mounted shear wave sensor produces a much sharper and shorter response to acoustic emission than does the surface mounted longitudinal sensors. This is significant to detection resolution and to source location accuracy.
3. The surface mounted longitudinal sensor is not suitable for use with cavitation noise present.
4. The quality of response obtained from edge mounted shear sensors on a block attached to the specimen surface demonstrates one way of applying these sensors on a

practical basis to existing structures. The response is definitely not optimum, however, and better techniques are currently being sought.

An acoustic emission monitor system has been installed on the Containment System Experiment (CSE) simulator vessel to monitor for any growth of known laminar flaws in the heads. This system has been operated during hydrostatic pressure and nozzle jacking tests preliminary to use of the vessel in routine CSE tests. The system operated satisfactorily during these tests. None of the data analyzed to date shows any evidence of flaw growth. However, deformation of temporary blind flanges 8 to 10 ft from the sensors during hydrostatic testing was clearly detected. This is particularly significant because the signals from the flange deformation had to transmit across a bolted interface to reach the sensors.

Work Planned For Next Quarter

- Complete the basic study of improved emission signal detection techniques.
- Design and test a practical method of applying the edge mounted shear sensor in the field.
- Continue with an environmental test of high temperature transducer crystal materials.
- Modify the existing broadband solid state preamplifier to operate over a limited range of about 1.5 to 2.5 MHz.
- Determine the maximum range of emission signal detection using the edge mounted shear technique.
- Work cooperatively with Phillips Petroleum Company to obtain noise background measurements on a pressurized water power reactor.
- Issue topical reports on a) basic study of detection techniques, and b) test performed to demonstrate detection of acoustic emission in the presence of hydraulic noise.

References

1. P. H. Hutton. Nuclear Reactor System Noise Analysis, Dresden I Reactor, Commonwealth Edison Co., BNWL-867.
Pacific Northwest Laboratory, Richland, Washington,
September 1968.

4. ENVIRONMENTAL STUDIES

R. F. Foster

Contributions to this section were not received in time to be included in this report.

5. FIXATION OF RADIOACTIVE RESIDUES

SPRAY SOLIDIFICATION

Flowsheets for Processing PW-4m in the Spray Solidifier -

J. E. Mendel

A large number of nonradioactive PW-4m melts of the borosilicate type were prepared. Although the laboratory investigation is continuing, present indications are that a phosphate, rather than borosilicate flowsheet, will be favored for processing PW-4m in the spray solidifier. Some reasons for this conclusion are:

- Rare earth precipitates were found in most of the PW-4m borosilicate melts.
- Feed preparation and handling are more difficult for the borosilicate than for the phosphate flowsheet.
- Borosilicate products were more leachable than phosphate products.

The range of additives used in preparing the PW-4m borosilicate melts included:

<u>Additive</u>	<u>Concentration Range, $\underline{M}^{(a)}$</u>
Boron	3.0-9.0
Silicon	0-2.0
Alkali metal (Li, Na, or K)	0-3.0
Alkaline earth (Ca or Ba)	0-1.6
Other cations (Al, Fe, or Mo)	0-0.6

a. Moles per liter at 378 liter/tonne

It is noteworthy that all of the 69 compositions tested produced mobile melts at temperatures ranging from 850 to 1100 °C. In contrast, experience with the phosphate system has shown that comparatively greater restrictions on composition are necessary in order to obtain fluid melts in this temperature range.

A basic problem with the PW-4m borosilicate melts was non-homogeneity in the form of a precipitate containing rare earths. PW-4m contains 0.274M fission product rare earths. The precipitate was heavier than the bulk glassy phase and collected at the bottom of the melt. Segregation of this rare earth enriched phase in the waste storage containers would probably be undesirable, and efforts to define a PW-4m borosilicate composition range which totally avoids the precipitate were unsuccessful.

Feed solutions were prepared by dissolving the required amount of B_2O_3 in the required amount of alkali metal hydroxide and adding PW-4m to this solution. Silicon, when used, was added as SiO_2 flour and the other cations as nitrates. The feed solutions were viscous and somewhat gelatinous and proved difficult to handle in the engineering-scale equipment.

The flowsheet that will be used for the first phosphate-fluxed PW-4m spray solidifier runs will be related to the PW-1 flowsheet, which has proven quite troublefree. The additives will be 1.2M Fe, 0.75M Li, 0.75M Na and 2.70M PO_4 (where M equals moles per liter of PW-4m at 378 liter/tonne). Although the melt properties are similar to the PW-1 product, the fission product content is higher, 24.7 wt% in PW-4m product versus 17.8 wt% in PW-1 product.

Ruthenium Volatility Studies - D. W. Brown

One series of feeds was calcined at 375 °C to determine the effect of the neutralization of the feed on ruthenium volatility. A PW-2 spray solidifier flowsheet feed with 0.016M Ru was used and the acid was neutralized with sodium hydroxide. Figure 5.1 is a summary of the data. A continued increase in ruthenium retention occurs as the percent of nitric acid neutralized is increased and if 100% excess sodium hydroxide is added (200% neutralized), less than 1% of the ruthenium is volatilized.

FIGURE 5.1. Neutralization of PW-2 Acid with Sodium Hydroxide and Its Effect on Ruthenium Volatility

A second set of PW-2 spray solidifier feeds, 0.008M Ru, were calcined to determine the effect of sugar in the feed solution on ruthenium volatility. This method also successfully reduced the ruthenium volatility as shown by the following data:

<u>Sugar in Feed, M</u>	<u>Ruthenium Volatilized, %</u>
0.0	23
0.26	14
0.51	2.7

No direct comparison can be made between the two sets of data because the two base-feed solutions were of different ruthenium concentrations. Additional tests are planned so that a comparison can be made.

Corrosion Behavior of Materials for High Temperature
Melt Processing - R. F. Maness

The corrosion behavior, as well as the thermal and mechanical shock resistance, of Haynes Metal Ceramic LT-1 (77Cr-23 Al₂O₃) and of chromium (both deposited by flame spraying) were determined during the formation of phosphate glass and spray solidifier type PW-1 melts. Melts were formed at 1000 °C in containers of the two materials. Corrosion after 100 exposure hours was not excessive as evidenced by visual observation and the containers, partially filled with melt, retained their integrity when cycled between 1000 °C and room temperature. However, corrosion of both materials was severe at 1150 °C. Specimens of Hastelloy D* and Duriron** corroded severely when exposed to PW-1 spray solidifier melt at 1050 °C.

* *Hastelloy D is a registered trademark of the Union Carbide Corp.*

** *Duriron is a registered trademark of the Duriron Co.*

A 50 Cr-50 Ni alloy specimen exposed to PW-1 phosphate glass melt at 900 °C for 80 hr corroded at a rate of about 240 mils/year. PW-1 and PW-2 spray calciner melts both corroded the alloy at a rate of about 100 mils/year. Corrosion rates of the 50 Cr-50 Ni alloy at 800 °C in PW-1 spray solidifier and phosphate glass type melts, and PW-2 spray solidifier melt, were 7, 17, and 60 mils/year, respectively, as determined in a 145 hr test. The alloy may be useful as a material of construction for thermocouple wells in containers containing melts with molten cores. One-eighth inch thick specimens of 310 Stainless Steel, Hastelloy X*, and Inconel 600 ** exposed to PW-2 spray calciner melt at 900 °C were consumed after 165 exposure hours.

Development Spray Solidification Runs - J. D. Moore

Three runs with the cold semiworks spray solidifier were made during the quarter. Run DSC-19 was made to produce 100 lb of simulated PW-1 calcine for Oak Ridge National Laboratory. The calcine will be used as base material for development of high thermal conductivity glass for solidification of fast reactor fuel reprocessing wastes. Runs DSC-20 and 21 tested simulated PW-4m with borate flux in an attempt to develop a feed composition from this waste type suitable for spray solidifier operation. PW-4m, containing 1.5M Li and B₂O₃ added as a solid flux to the melter in an equivalent concentration of 3.0M (based on 378 liter/tonne feed), provides a workable feed which forms a glass product containing greater than 95% of the feed ruthenium.

* *Hastelloy X is a registered trademark of the Union Carbide Corp.*

** *Inconel 600 is a registered trademark of the International Nickel Co., Inc.*

A. Run DSC-19

The feed rate and concentration were varied in DSC-19. For the first 9 hr of the run, an average rate of 15 liters/hr was maintained with PW-1 feed concentrated to 303 liters/tonne (see Table 5.1). For the last 7.7 hr, 227 liter/tonne feed was spray-dried at an average rate of 18 liters/hr to hasten completion of the run. The calciner internal temperatures indicated very smooth operation. With the calciner furnace temperature at 700 °C, the internal temperatures were 15 to 20 °C cooler at 18 liters/hr than at 15 liters/hr. The

TABLE 5.1. Summary of Development Spray Solidification Runs

	DSC-19		DSC-20	DSC-21
	Part 1	Part 2		
Feed Type	PW-1	PW-1	PW-4m	PW-4m
Feed Concentration, liters/tonne	303	227	378	447
Additives	none	none	Li ⁺ , SiO ₂ to liquid B ₂ O ₃ to melter	Li ⁺ , B ₂ O ₃ to liquid
Feed Consumed, liters	136	137	48	10 (estimate)
Average Feed Rate, liters/hr	15	18	11.4	11 (estimate)
Calciner Furnace Temperature, °C	700	700	700	700
Calciner Vacuum, in. of water	5	5	7	10
Atomizing Gas	Air	Air	Air	Air
Atomizing Gas Flow, scfm	6	5.6	6	6.5
Filter Blowback Pressure, psig	40	40	30	30
Filter Pressure Drop, End of Run, in. of water	5.75	6.5	4.3	2.5
Melter Furnace Temperature, °C	--	--	1000-1100	--
Product Receiver Furnace Temperature °C	--	--	800	800
Product Collected, kg	23.4	31.1	16.8	2 (estimate)
Product Density, grams/cm ³	--	--	2.6	3.2
Particulate DF (Feed to Condensate)	600	500	360	3300
Ruthenium Retention in Product, %	<5	<5	>95	not det.

equilibrium calciner filter pressure drop was 13% higher (5.75 vs 6.5 in. of water) at the higher feed rate. Particulate decontamination factors (DF) remained relatively constant from 500 to 600 throughout the run.

Calcine product was collected in an unheated stainless steel drum connected to the calciner outlet. A total of 120 lb of fine, dry homogeneous, brown powder was produced.

The calciner internal walls were relatively clean with no deposits exceeding 1/8 in. thickness.

A ruthenium material balance gave a poor recovery as less than 5% of the feed ruthenium was found in the product and only 13% was found in the condensate and scrub solution.

B. Runs DSC-20 and 21

Runs DSC-20 and 21 were made to compare the spray solidifier performance with PW-4m feed with B_2O_3 flux added as a solid to the melter during DSC-20 and added to the liquid feed to the calciner during DSC-21. Feed for DSC-20 (see Table 5.2) was simulated 378 liter/tonne PW-4m with 1.5M Li and 0.8M SiO_2 added to the liquid feed, and with 3.0M B_2O_3 added in crystalline form to the melter. For DSC-21, the silica was omitted since it caused severe wear to the feed pump bearings in the previous run. Its presence in the feed was to improve product solubility; however, it was found to have had little effect in this respect. The B_2O_3 was added to the liquid feed in DSC-21 but it did not completely dissolve and a heavy crystalline precipitate formed when the feed was cooled below 50 °C.

The solid B_2O_3 addition mode proved to be more successful than the liquid addition. In DSC-21, with the flux in the liquid feed, the feed formed a thick paste in the storage tank after about 55 min of spray solidifier operation. The paste

TABLE 5.2. Feed Compositions for Development of Spray Solidifier Runs

Basis: 378 liters/tonne Concentration

	DSC-19	DSC-20	DSC-21
Waste Type	PW-1	PW-4m	PW-4m
H ⁺	3.52	4.81	4.81
Fe ⁺³	0.959M	0.124	0.124
Cr ⁺³	0.012	0.012	0.012
Ni ⁺²	0.005	0.008	0.008
Al ⁺³	0.001	0.001	0.001
ZrO ⁺²	0.065	0.143	0.143
Na ⁺	0.138	0.342	0.342
SiO ⁻²	0.010	0.010	0.010
NO ₃ ⁻	7.5	6.66	6.66
PO ₄ ⁻²	0.003	0.003	0.003
MoO ₄ ⁻²	0.065	0.13	0.13
Sr ⁺²	0.0155	0.036	0.036
Ba ⁺²	0.0195	0.041	0.041
Rb + Cs (used K ⁺)	0.0416	0.092	0.092
(Y + RE) ⁺³	0.12	0.274	0.274
Ru ⁺⁴	0.0032	0.0082	0.0082
Rh (used Co ⁺²)	0.0074	0.013	0.013
Ag ⁺	0.0008(used Cu ⁺²)	0.0016	0.0016
Cd ⁺²	0.0008(used Cu ⁺²)	0.0025	0.0025
Te (used SO ₄ ⁻²)	0.0064	0.014	0.014
Tc	0.014(used Mn ⁺²)	0.031(used MoO ₄ ⁻²)	0.031(used MoO ₄ ⁻²)
Pd(used Ni ⁺²)	0.0172	0.043	0.043
<u>Additives</u>			
LiOH		1.258	1.258
SiO ₂		0.8	--
B ₂ O ₃		3.0(solid to melter)	3.0

was formed when the feed was gradually cooled to 40 °C while circulating through the pump loop back to the tank. The handling problems with this feed make it infeasible for further testing at this time.

Silica was the only insoluble precipitate in the feed for DSC-20 and without it the feed would probably have run smoothly. As the silica began to wear the pump bearings, the feed line pressure oscillated between 25 and 45 psig (normal setting was 35 psig). These pressure fluctuations produced large droplets in

the feed atomization pattern which formed calcine deposits up to 3/4 in. thick in the calciner cone below the spray nozzle. Sixty-mesh B_2O_3 crystals were added to the melter through a nozzle in the calciner cone. Moisture condensation in the crystals at the nozzle outlet plugged the addition port after 2 hr. For the remaining 2 hr of the run, solids were added manually through a port in the melter top. The solids plug could have been prevented by continuously purging the solids addition line with heated air.

The calciner filter pressure drop was relatively low in both runs, although longer duration runs would have yielded more useful data.

Melt from DSC-20 was a homogeneous dark olive-green glass with a density of 2.6 g/cm^3 and a melting point of 850°C . Product from DSC-21 contained small light-colored inclusions. It melted at 950°C and had a density of 3.2 g/cm^3 . The higher density and melting point, and crystalline structure, were due to a borate deficiency in the melt. These occurred because of borate feed handling problems described above.

Greater than 95% of the feed ruthenium was retained in the melt from DSC-20. Since DSC-21 was so short, no ruthenium analytical data were obtained.

New Freeze Valve Cooling Coil Assembly - J. D. Kaser

A new method has been developed to control the discharge of melt from the spray solidifier melter. The melt in the valves is solidified by air cooling coils. To discharge the melt, the cooling air is turned off to either the weir overflow valve or the drain valve and the heat from the melter furnace thaws the material in the valves. A complete cooling coil assembly has been fabricated, tested, and installed in B-Cell.

Initial operating experience, including its successful use in WSEP Run SS-5, has shown that this method bears high promise as a simple and reliable replacement for the previously used electric heaters. The electric heater system had experienced an excessive failure rate and consequently this change should provide a significant improvement in the reliability and operability of the solidifier system.

Figure 5.2 shows a top view of the cooling coil assembly. The coils were wound from 1/4 in. outside diameter Hastelloy X* tubing. The coils are as long as possible without touching any part of the melter, with the valve tips extending about 1 in. below the coils. The cooling coil for the drain valve is 4 in. long and the coil for the overflow weir valve is 2 in. long. The coils have an inside diameter of 1 in. and are wound without gaps between turns. The cooling air flows from the bottom to the top of the coils and is then exhausted from the assembly.

The coils are not intended to freeze the melt in the valves while the melter is draining rapidly. Rather, the coils seal the valves after flow has stopped. At the beginning of a run, the slow trickle of melt into the cooled valves forms the initial seal.

The outside of one coil is insulated with 1/2 in. thick Cerafelt** covered with platinum foil. This insulation reduces the interaction between cooling coils. Without insulation, cooling air flowing in one coil may freeze both valves. The coil assembly is surrounded by 1/2 to 1 in. thick Cerafelt to minimize heat loss from the valves when the cooling air is off. Insulation below the coils (not shown) is supported by the stainless

* Haynes Stellite Corporation

** Johns-Manville, Inc.

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FIGURE 5.2. Top View of Freeze Valve Cooling Coil Assembly

steel plate which is shown. The melt falls through the large holes in the plate. To further reduce heat loss the entire assembly was lined with platinum foil (not shown).

Experience has revealed that molten waste will rapidly corrode Hastelloy X at the operating temperatures of the melter (900 to 1200 °C) if metal and melt come in contact. Furthermore, the platinum valves may be attacked if the valves, the melt, and the alloy coil are all in contact with each other. To prevent such contact, a platinum wire cage is suspended inside each coil.

After the assembly was fabricated, the temperatures at the valve tips were measured both inside and outside the valve at various cooling air rates. The thermocouples measuring the temperatures outside the valves were permanently installed for control purposes. Tests were run at two temperatures: 920 °C, corresponding to PW-2 operation, and 1150 °C, corresponding to PW-1 and PW-4 operation. The test results are summarized in Table 5.3. These tests show the decreasing valve temperatures with increasing cooling air flow, and the moderate interaction between coils. The valve tips must be above 750 °C to thaw PW-2 waste and above 925 °C to thaw PW-1 waste. In actual operation (WSEP Run SS-5), the valve tip temperature ran about 50 °C higher than the test data at the same operating conditions.

This difference may be attributed to the platinum foil lining and to the fact that during the tests the coils radiated heat through the drain holes at the bottom of the assembly to a cold laboratory floor, while in actual operation the coils see the inside of the receiver pot which is about 700 °C.

TABLE 5.3. Final Tests of Freeze Valve Cooling Coil Assembly
(Assembly not Lined with Platinum Foil)

Furnace Temp, °C	Drain Valve Tip, °C		Weir Tip, °C		Air Flow, SCFM	
	Inside	Outside	Inside	Outside	Drain Valve	Weir
920	750	750	750	760	0	0
	730	730	675	690	0	0.7
	725	725	645	665	0	1.3
	670	670	730	<600	0.7	0
	<600	600	705	<600	1.3	0
1150	980	990	975	985	0	0
	960	960	875	900	0	0.7
	950	950	845	885	0	1.3
	900	920	950	960	0.7	0
	800	850	920	930	1.3	0

RADIOACTIVE DEMONSTRATION

Spray Solidification Run SS-4 - W. R. Bond, M. R. Schwab,
and J. L. McElroy

The fourth radioactive demonstration of the spray solidification process was successfully completed with Run SS-4, WSEP Run 16. During SS-4, 500 liters of PW-2 waste at 560 liters/tonne were reduced to 56 liters of calcine in 40.5 hr. The 1,300,000 Ci of fission product activity (including 3,500 Ci of ruthenium) in the solidified waste produced an internal power generation of about 5400 W. The centerline temperature of the 8 in. diameter mild steel receiver in the air-cooled furnace was 510 °C with a pot wall temperature of 290 °C. The 220 °C temperature drop gave an effective thermal conductivity of $1.1 \text{ W}/(\text{m}^2)(^\circ\text{C}/\text{m})$ [$0.66 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F}/\text{ft})$] for the solidified waste product. Table 5.4 presents a summary of the data for Run SS-4.

The operation of the spray solidification systems was quite smooth. The principal process factors (atomization, filter operation, flow control) performed very satisfactorily. A new two-filter blowback assembly (two filters blown back simultaneously) produced much lower pressure surges than observed in previous runs with a three-filter blowback arrangement. Air blowback was used during Run SS-4 instead of steam due to a

TABLE 5.4. Summary Data for WSEP Spray Solidifier Runs

Operational Data	SS-4	SS-5
Solidifier Run/WSEP Run	4/16	5/17
Solidifier	Spray	Spray
Date	9/68	10/68
Operating Mode	A	A
Feed Type	PW-2 ^(a)	PW-1
Feed, liters/tonne	560	446
Pot Diameter, in.	8	8
Pot Material	Mild Stl.	Mild Stl.
Total Run Feed Time, hr	40.5	53.2
		3.5
Average Feed Rate, liters/hr	12.3	16.7
Total Feed, liters	500	887
Total Feed, tonne equivalent	0.89	2.0
Feed: Solid Volume Ratio	8.9	13.3
Feed Solid Volume Ratio at 378 liters/ tonne Basis	6.0	11.4
Filter, ΔP , in. of H ₂ O	7.0 ^(b)	6.0
<u>Problems</u>		
SS-4 Weld on vibrator mounting plate failed; new unit installed and run continued.		
SS-5 Power lead to melter furnace lost insulation and began arcing; insulation replaced with 3 1/2 hr delay in run.		
<u>Filled Pot Data</u>		
Total Curies in Feed to Pot	1,300,000 ^(c)	2,000,000 ^(d)
Total Curies of Ru in Feed to Pot	3500 ^(c)	17,000 ^(d)
Solid in Pot, liters	56	66
Solid in Pot, kg	160 ^(e)	214 ^(f)
Solid in Pot, liters/tonne	63	33
Heat in Pot, W ^(g)	5400	8200
Equivalent Aging Time, year	0.8	1.1
$\bar{C}$ Temp in Air-Cooled Furnace, °C	510	633
$\bar{C}$ to Wall Δ Temp., °C	218	278
Effective Thermal Conductivity, W/(m ²)(°C/m) [Btu/(hr)(ft ²)(°F)]	1.1 [0.66]	1.1 [0.64]
Welded Lid	Yes	Yes
Radiation Reading, R/hr at 6 in.	3.8 × 10 ⁴	—

^{a.} See Table 5.2 for feed composition.
^{b.} Approximately 3 in. of ΔP was due to locked open blowback solenoid.
^{c.} By melt analysis
^{d.} By feed analysis
^{e.} Calculated from lab density of 2.84 kg/liter and rodded volume in pot.
^{f.} Calculated from lab density of 3.24 kg/liter and rodded volume in pot.
^{g.} By feed volume ratio; actual value pending pot calorimetry data.

continuous air blowback on one filter bank caused by a solenoid valve becoming locked open. A new vent line between the process condenser and the auxiliary evaporator provided ample vacuum in the solidifier even with the continued air blowback.

The only difficulty which occurred during SS-4 was a failure of the mounting of the vibrator on the spray calciner (the welds holding the base of the mounting to the spray calciner broke loose). This necessitated an interruption of the run while a floating-type vibrator was fabricated and placed on the calciner.

The batch dumping of the molten product into the unheated receiving pot was completed without stalagmite formation. The melter was dumped about every 3 hr by heating the freeze valve with a propane torch. An interesting feature of the PW-2 product was the sensitivity of the flux-to-feed addition ratio on the sulfate volatility. Each time the melter was dumped, sulfate fumes were observed coming from the flowing product. The flux (sodium hexametaphosphate) was added batchwise as a solid to the melter every half hour to maintain a flux to feed ratio of 87 g NaPO_3 /liter of feed at 555 liters/tonne. The sulfate fumes became very dense each time the flux-to-feed ratio was allowed to exceed the nominal 87 g/liter, viz., up to 90 g/liter. Conversely, the least sulfate fuming occurred for a flux-to-feed ratio of 85 g/liter. While the flux is required to lower the melting point of the calcine, the melting point did not appear to be as sensitive to the flux-to-feed ratio as was the sulfate volatility.

The surface of the solid product was smooth and level on inspection of the receiving pot following the run.

The feed composition for Run SS-4 is given in Table 5.5. The feed differed from a standard PW-2 waste formulation in the amount of aluminum present. The excess of aluminum in the Purex 1WW waste solution used to prepare the feed was used for 13% of the iron requirements in the PW-2 feed. The aluminum replaced 0.059M Fe of the total iron requirement of 0.475M (including 0.0288M Fe substituted for ruthenium) at 378 liters/tonne.

The distribution of the feed radioactivity following Run SS-4 is given in Table 5.6. About 11% of the radioruthenium fed to the solidifier was found in the auxiliaries following the run. While the 11% ruthenium volatility value agreed with stream sample data from the process condenser, only 23% additional ruthenium was found (by melt analysis) in the product.

TABLE 5.5. PW-2 Feed Composition for Run SS-4

Component	Molarity, M ^(a)	Lab Analysis, M ^(a)
H ⁺	---	5.5
Fe ⁺³	0.445	0.415 ⁽⁴⁾
Cr ⁺³	0.024	
Ni ⁺²	0.01	
Al ⁺³	0.001	
Na ⁺¹	0.93	1.02
U ⁺⁶	0.01	
NO ₂ ⁻¹ -NO ₃ ⁻¹	5.37	7.14 ^(b)
SO ₄ ⁻²	0.87	0.66 ^(c)
PO ₄ ⁻³	0.006	
SiO ₃ ⁻²	0.01	
Cs ⁺ (added K ⁺)	0.042	
Sr ⁺²	0.0155	
Ba ⁺²	0.0195	
(Y+RE) ⁺³	0.120	0.23 ^(g)
ZrO ⁺²	0.065	
MoO ₄ ⁻² + Tc as MoO ₄ ⁻²	0.079	
Ru ⁺⁴	0.0032	0.0020
Ru ⁺⁴ (added Fe ⁺³)	0.0288	(d)
Rh ⁺² (added Co ⁺²)	0.0074	
Pd ⁺² (added Ni ⁺²)	0.0172	
Ag ⁺¹ + Cd ⁺² (added Cu ⁺²)	0.0024	
TeO ₄ ⁻² (added SO ₄ ⁻²)	0.0064	
<u>Additives to Feed</u>		
Al ⁺³	0.25	0.31 ^(d)
Ca ⁺²	0.60	
Li ⁺¹ (as hydroxide)	1.17	
Na ⁺¹ (added as NaPO ₃ solid directly to melter)	1.27	
PO ₄ ⁻³ (total including 1.61 as H ₃ PO ₄)	1.61	1.58
<u>Final Feed</u>		
Volume, liter		569
Waste Equivalents		1.01
$\sum M_T^+$ for Feed (for melt; includes NaPO ₃ additive)	6.11 (7.38)	
$\sum M_T^+$ / P for feed (for melt; includes NaPO ₃ additive)	3.80 (2.56)	
Sp Gr @ 25 °C		1.36
% Solids		30
Total Radioactivity, Ci/liter ^(e)		3200
Radioruthenium, Ci/liter ^(e)		31
Total Heat, W/liter ^(f)		11

a. Molarities are at 378 liters/tonne basis.

b. Includes nitrate from additives

c. Presence of insoluble sulfates (probably calcium sulfate) gives lower value than known to be actually present.

d. Excess aluminum in IWW waste replaces 0.059M Fe in the total iron requirement of 0.475M (including 0.0288M Fe substituted for ruthenium).

e. By feed analysis. Radioactivity in product was determined by analysis of the product.

f. As determined by calorimetry measurement in feed tank.

g. Rare earth analysis has history of being off as much as a factor of 2.

The remaining 66% of the radioruthenium fed to the solidifier was not found although additional laboratory analyses are pending.

Instantaneous decontamination factors were determined for ruthenium across the WSEP auxiliary equipment. Instantaneous ruthenium DF's for the evaporator to the evaporator overheads ranged from 500 to 2500. Instantaneous ruthenium DF's for the fractionator to the fractionator overheads ranged from 10 to 30.

Spray Solidification Run SS-5 - W. R. Bond and J. N. Hartley

The seventeenth WSEP pilot plant demonstration of the solidification of high level radioactive waste was successfully completed with the fifth 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 power density of 125 W/liter (8200 W total based on feed analytical data) in an 8 in. diameter 304-L Stainless Steel melt receiver. The steady-state pot centerline temperature in the air-cooled furnace was 633 °C and the centerline-to-wall temperature difference was 278 °C. These values gave an effective thermal conductivity of $1.1 \text{ W/(m}^2)(^{\circ}\text{C/m)}$ [$0.64 \text{ Btu/(hr)(ft}^2)(^{\circ}\text{F/ft)}$] for the ceramic product. Preliminary data for Run SS-5 is presented in Table 5.6.

TABLE 5.6. Activity Distribution for Run SS-4

	Fraction of Radioactivity in Condensate				
	Ru	CePr	Cs	ZrNb	Total less Ru
Through Process Condenser (E-111)	1.1×10^{-1}	1.2×10^{-3}	1.4×10^{-2}	2.5×10^{-3}	1.2×10^{-3}
Through Evaporator Condenser (E-113)	7.4×10^{-4}	8.5×10^{-7}	5.1×10^{-5}	1.2×10^{-6}	9.4×10^{-7}
Through Fractionator Condenser (E-115)	2.7×10^{-5}	3.1×10^{-9}	3.9×10^{-8}	----	3.1×10^{-9}
Off-Gas to Scrubber	3.0×10^{-4}	7.4×10^{-10}	1.1×10^{-7}	----	1.5×10^{-9}
Off-Gas to Stack	----	----	----	----	1.0×10^{-12}
Ratio (Fract. Cond.) Activity to 10CFR20	77,000	570	14	----	----

The calciner was fed 887 liters of adjusted sulfate-free PW-1 feed (additives of 0.5M LiOH, 0.5M NaOH, and 1.71M H_3PO_4 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 contained 2,000,000 Ci of radioactivity, including 17,000 Ci of radioactive ruthenium, and was reduced in volume by a factor of 13.3 (11.4 on a 378 liters/tonne basis).

A new freeze valve cooling assembly (see Engineering Studies - New Freeze Valve Cooling Coil Assembly) was used throughout the run. The melter was batch-dumped through the drain valve eleven times (about every 5 hr). The drain valve began to flow about 2 min after the air was turned off to the cooling coils around the valve. The use of air-cooled coils was an effective means of sealing the drain valve following each batch dump of the melt to the receiver.

Additional information on Run SS-5 will be reported in the next quarterly report.

Off-Gas Filter Failure - W. R. Bond

The spray solidifier off-gas filters were removed from the unit following Run SS-3 for inspection as an appreciable quantity of calcine (11%) passed the filters during the run. It was found that two of the fifteen sintered powder metal (316-L SST) filters had failed. Both failures occurred just below the top circumferential weld (manufacturer's weld) which joins the porous metal to a solid collar. The collar connects the filters to venturis through which the off-gases exit, and back through which a pulse of superheated steam is directed periodically for filter cleaning. The filters failed after 400 hr of service (feed-on time). They were visually inspected after the first 300 hr and appeared in good condition at that time. One filter had a 2 in. long by 1/32 in. wide crack in the porous metal

along the circumference in the weld-affected zone. A 1 to 1 1/2 in. section of porous metal was missing from the top of the second filter. The porous metal 1 to 2 in. directly below this failure appeared pitted as though numerous metal particles were missing. A sample of the porous metal (Figure 5.3) taken in this area was very crumbly and possessed no ductility. Another sample (Figure 5.4) taken 4 to 5 in. below this point was not pitted and possessed some ductility. Both samples were sent to Radiometallurgy for metallographic examination. Although the examination revealed that an appreciable amount of metal particles were missing from the sample taken near the failure, signs of corrosion of the remaining particles were no more pronounced than on the other sample which was considerably more intact. To date, a combination of mechanical fatigue and corrosion is suspected as the cause of the failure. Fatigue of the top of the filter could result from a pivoting action about the porous metal to solid metal weld caused by vibration. A filter manufacturer was contacted and it was recommended that a perforated support plate be installed on the inside of the filters to minimize the mechanical stressing of the porous metal. Materials of construction that may be more resistant to corrosion than 316-L SST are also being considered. The replacement set of filters now in the calciner was an existing spare set and as such is similar in most respects to the original filters.

Computer Data Logging

A G.E. 412 computer has been used successfully during the report period by Engineering Demonstration personnel to assist in the execution of experimental run plans and in the post-run evaluation of these experiments. The computer has been valuable in calculating and logging routine data associated with both pre-start-up activities and normal operating periods when operating personnel time was more beneficially used for other activities.

Neg E6470 2.5X
Sample as Received

Neg E6478 25X
*Cross Section of
 Sample as Etched*

Neg E6480 125X
*Cross Section of
 Sample as Etched*

FIGURE 5.3. *Sample of Porous Metal Filter Taken at the Failed Area*

Neg E6471 2.5X
Sample as Received

Neg E6485 25X
Cross Section of
Sample as Etched

Neg E6486 125X
Cross Section of
Sample as Etched

FIGURE 5.4. *Sample of Porous Metal Filter Taken 4 to 5 Inches*
Below the Failed Area

The 412 has been programmed to continuously monitor many of the process sensors of the WSEP facility, calculating and logging specific values of interest and alarming out-of-limit conditions. Data were logged either at 15 min intervals or on demand with typewriters or paper tape perforators. Alarming was accomplished with typewritten messages. Typewriter logged data were expressed in convenient engineering units that were directly usable by operators for controlling the progress of the experiment or by engineers evaluating the experiment. Additional data were logged with paper tape perforators for later evaluation by engineering personnel using electronic data processing facilities. Using this technique, certain data were machine plotted, eliminating the time consuming process of manually calculating and hand plotting of these data.

Typical logged data included:

1. Feed solution specific gravity, volume and dropout rate.
2. Auxiliary train volumes, specific gravities, temperatures, acid concentrations and flow rates for acid material balance calculations.
3. Furnace power consumption.
4. Material balance calculations.
5. Clock time and elapsed run time.

PHOSPHATE GLASS SOLIDIFICATION

Zircaloy-2 Corrosion - R. F. Maness

Zircaloy-2 was evaluated as a material of construction for containing PW-1 and PW-2 phosphate glass feed solutions boiling at 120, 130 and 140 °C. Corrosion rates ranged from 0.01 to 2.5 mils/month. Corrosion rates of Zircaloy-2 in boiling melter condensate ($4.2\text{M H}_2\text{SO}_4 - 2.1\text{M HNO}_3 - 0.2\text{M H}_3\text{PO}_4$) and in melter condensate plus PW-2 feed solution were 0.01 and 0.4 mils/month, respectively. These rates were appreciably lower than those found for any common material of construction evaluated to date.

STUDIES ON AUXILIARIES TO SOLIDIFICATION SYSTEMSReaction Kinetics of Sucrose Added to the Evaporator -

P. C. Owzarski

Sucrose will be added to the WSEP evaporator to reduce ruthenium volatility by producing reducing oxides of nitrogen. It is necessary to understand the sucrose-HNO₃ reaction to predict transient behavior of the evaporator. Existing batch reaction data were examined using the following best fitting kinetic models:

$$\frac{d[\text{carbon}]}{dt} = -k [\text{carbon}][\text{HNO}_3]$$

$$\frac{d[\text{HNO}_3]}{dt} = -\alpha k [\text{carbon}][\text{HNO}_3]$$

The constants k and α are dependent on temperature and concentration of various ions (Fe⁺³ most notably). The following table was computed from 24 hr batch denitration data at 100 °C. The rate constants are based on carbon concentration (moles sucrose/liter) and α is the number of moles of HNO₃ destroyed per mole of sucrose reacted.

<u>[Fe⁺³] moles/liter</u>	<u>α</u>	<u>k, liter/mole min</u>	<u>αk</u>
0	12	0.00070	0.0083
0.1	15	0.00057	0.0086
0.5	18	0.00087	0.016
0.8	20	0.0026	0.052
1.0	20	0.0028	0.056

EVALUATION OF SOLIDIFIED WASTE PRODUCTS

Study of the High Temperature Storage of Solidified Waste Products Containing Sulfate - D. W. Brown

This experiment is designed to determine whether the sulfate in solidified waste products containing sulfate will decompose under conditions of high temperature storage and result in excessive vessel pressurization.

A vessel 12 in. in diameter and 26 in. tall was filled 22.5 in. deep with Spray Solidification PW-2 (sulfate-containing) melt. This was heated at the center of the ceramic product.

An equilibrium temperature of about 500 °C at the ceramic-heater interface was held for nine days and no pressurization due to sulfate decomposition was observed. When a temperature of 600 °C was attempted, the heater burned out. A second heater was installed in the same manner and it too burned out on attempting to reach 600 °C. In both cases, there was a fifteen mil gap between the heater and the platinum sheath. A third heater, with the platinum sheath drawn onto the heater, has been installed in the vessel and this should allow a temperature of 700 °C at the ceramic-heater interface.

Product Characterization - Continuation of Laboratory Evaluation of PW-2 Phosphate Glass Samples - P. L. Farnsworth and W. L. Mularz

Thermal Diffusivity

Optimum sample geometry for thermal diffusivity measurements is 0.5 in. diameter and 0.027 in. thick. One side of

each sample is vapor coated with a layer of chromium and gold for electrical and thermal contact of the thermocouple. The vapor-deposited films adhere to the substrate during heating without any detrimental effect on the measurement.

Preliminary results show that the thermal diffusivities of the samples do not vary greatly with sample history and temperature and are of a low value (see Table 5.7).

Thermal Conductivity

The thermal conductivity of Samples 6, 9, and 10 (see Table 5.7) is being measured by a comparative technique using a Thermo-Physics Corp. Model TC-2000 thermal conductivity apparatus. The apparatus has been calibrated and measurements are in progress. Other samples are being machined.

Dispersibility

A prototype sample (0.1 in. diameter $\times$ 0.50 in. long) was shattered by a 2.9 lb weight with a blunted steel tip (0.12 in. diameter) dropped from a height of 5 ft. The particles were sieved and classified as to size and weight. Results are shown in Table 5.8 and Figure 5.5.

The quantity of sample passing through a 230 mesh screen is less than 0.1%. Characterization will include only particles greater than 325 mesh.

Additional samples have been prepared and will be evaluated. Their geometry is 0.75 in. diameter by 0.50 in. long.

Coefficient of Linear Thermal Expansion (CLTE)

The linear thermal expansion of eleven samples has been measured from 200 to 500 °C. The results are tabulated in

TABLE 5.7. Sample History and Characterization of PW-2 Phosphate Glass Samples

Sample No.	Processing Temperature, °C	Time of Process, hr	Cooling Rate, °C/hr	Temperature for 8 Weeks, °C	% Solubility in H ₂ O	Density, g/ml	Porosity, %	Appearance after CLTE (a)	CLTE x 10 ⁻⁶ in./in./°C			
									200 °C	300 °C	400 °C	500 °C
4	1000	1	30	100	5.57	2.94	0	some glassiness	9.8	10.6	12.8	
5	1000	1	100	100	4.46	2.89	1.0					
6	1000	25	30	100	5.07	2.81	0	slightly glassy one end	9.1	10.8	11.6	
11	1000	25	100	100	1.48	2.89	0					
7	1000	1	30	600	41.35	2.99	2.2	heavily glassed	5.8	12.6	9.4	2.3
8	1000	1	100	600	33.84	2.98	3.2	heavily glassed	6.9	10.7	9.7	8.0
9	1000	25	30	600	39.27	2.98	2.4	some glassiness porous	7.1	8.5	4.6	7.6
16	1000	25	100	600	35.86	2.93	2.3	no change in appearance	10.1	13.8	14.2	11.6
12	1200	1	30	100	1.18	2.91	0	no change in appearance	10.3	9.1	7.5	
14	1200	1	100	100	1.39	2.92	0	glassy	9.4	9.8	7.1	
13	1200	25	30	100	1.63	2.90	0					
17	1200	25	100	100	2.18	2.91	0					
10	1200	1	30	600	1.93	2.88	0	no glassy appearance yellowish	9.8	11.0	12.1	11.6
2	1200	1	100	600	41.43	2.92	0	very glassy yellowish	10.8	11.8	14.4	11.7
15	1200	25	30	600	7.45	2.92	0					
3	1200	25	100	600	35.33	2.87	2.1	quite glassy one end	10.1	11.7	13.1	11.2

(a) Coefficient of Linear Thermal Expansion

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

<u>Sieve Opening^(a)</u>		
<u>in.</u>	<u>g</u>	<u>wt%</u>
0.312	4.588	21.3
0.223	6.9618	32.3
0.157	3.1942	14.8
0.111	2.5391	11.7
0.0787	1.5720	7.3
0.0555	0.8149	3.8
0.0394	0.4085	1.9
0.0278	0.3467	1.6
0.0197	0.2851	1.3
0.0139	0.1921	0.89
0.0098	0.1893	0.88
0.0070	0.1472	0.68
0.0049	0.1357	0.63
0.0035	0.1619	0.74
0.0025	0.0258	0.11
<0.0025	0.0030	0.01
Total Weight	21.565	99.94%

a. *The particles collected passed through a larger screen size opening and rest on the indicated sieve opening.*

FIGURE 5.5. *Dispersibility of PW-2 Phosphate Glass Samples - wt% Versus Size of Particles Collected*

Table 5.7 and plotted in Figures 5.6 and 5.7. The CLTE up to 300 °C is high and is comparable to iron, 12.5×10^{-6} in./in./°C. Although the composition is the same for all samples, their thermal histories vary and hence the CLTE varies. Normally, the CLTE is measured on annealed samples.

Heating of some of the samples caused gas evolution and a glassy appearance on the surface. Most notably affected were samples which were melted at 1000 °C and stored at 600 °C as indicated in Table 5.7.

Thermal Shock

A set of sieves will be used to classify particles which are produced by fracturing when the hot glass drops into a liquid medium at 0 °C. Because some samples are soluble in water, water will not be used as the quenching medium. Samples have been prepared and will be evaluated during the next quarter. Their geometries are 0.75 in. square by 0.12 in., 0.25 in., and 0.50 in. thick.

Crystallinity

All sixteen samples were mounted and polished. Their unetched microstructures are shown in Figure 5.8.

Melting and storage temperatures have the most significant effects upon the microstructure. Rate of cooling and time in the melt have less significant effect. The effect of storage time was not determined.

In samples melted at 1000 °C and 1200 °C and stored at 600 °C, the crystals are larger and more abundant than those

FIGURE 5.6. Coefficient of Linear Thermal Expansion of PW-2 Phosphate Glass Samples

FIGURE 5.7. Coefficient of Linear Thermal Expansion of PW-2 Phosphate Glass Samples

Neg 06899891

FIGURE 5.8. Microstructure of Phosphate Glasses
(All Samples Unetched 20X)

stored at 100 °C. Porosity is greater in samples melted at 1000 °C and stored at 600 °C than in samples melted at 1200 °C and stored at 600 °C. The samples heat treated at 600 °C were softer to abrasion than the samples which were heat treated at 100 °C.

Microstructure analysis and visual observation of Samples 2, 4, and 6 also revealed porosity in samples which were considered nonporous by mercury porosimeter measurements. Thin sections of the samples have been prepared and further microstructure analysis is continuing. Samples 2, 5, 8, and 14 will be examined for phase composition with electron beam microprobe spectroscopy and X-ray diffractometry.

PRODUCT MEASUREMENTS, TESTING AND STORAGE

Solids Storage Engineering Test Facility (SSETF) - D. H. Siemens and V. P. Kelly

Modification of A-Cell for SSETF has been virtually completed following installation of special in-cell mounting brackets, fabrication of the transfer mechanism and shielded ion chamber, and completion of most gallery items. Installation of the first-floor crane control outlet is not complete, but A-Cell startup will not be delayed by this item.

Design has been completed for gallery piping, electrical systems, and handling and transfer hardware for in-cell test pods and waste pots. Fabrication and installation of equipment and systems are expected to be completed in sufficient time to allow at least two weeks for remote design verification tests prior to the projected startup this year.

6. HEAVY SECTION STEEL TECHNOLOGY PROGRAM

IRRADIATION DAMAGE TO HEAVY-SECTION VESSEL STEELS -

J. A. Williams and C. W. Hunter

The objective of this program is to obtain a quantitative, engineering evaluation of the effects of irradiation on heavy-section, pressure vessel steels. Included in this broad objective are (1) the establishment of a suitable specimen and analytical approach for investigating the effects of irradiation on plane strain fracture toughness, (2) the development of valid correlations and surveillance methods for analyzing the service integrity of heavy-section vessels from fracture-mechanics specimen data, and (3) the determination of the effects of typical nuclear environments on possible microstructural embrittling reactions in the base metal, weld metal, and heat affected zones of candidate pressure vessel steels.

Investigations of Fracture Toughness of ASTM A 533 B Steel with the DCB Specimen

The size of specimens is recognized to be of major importance in irradiation experiments, both in terms of nuclear heating and neutron economy as well as test material consumption. A major task of the irradiation damage program was to compare the size effects of various fracture mechanics specimens to determine the optimum specimen for an irradiation program. The experimental work of this task at PNL has been primarily oriented towards the characterization of Double Cantilever Beam (DCB) specimen. The DCB type specimen offers some advantage over other types of specimens in that multiple fracture toughness measurements can be made, fracture arrest values can be determined, and fatigue crack preparation is unnecessary. Its major disadvantages (and lack of general acceptance) are related primarily to the limited degree of characterization of the

specimen, and more specifically, the limited knowledge as to the effect of side growths on plane strain fracture toughness.

The initial material for the PNL Irradiation Effects to Heavy Section Pressure Vessel Steels program was obtained from Plate 1 of A 533, Grade B, Class 1 steel procured for the HSST program. Heat treatment and characterization data of the plate is reported in Reference 1. Fracture initiation and fracture arrest measurements were made using the Double Cantilever Beam (DCB) specimen. A typical DCB specimen is shown in Figure 6.1. The specimen geometry used for this investigation for each size specimen was $B = h/2 = w/0.35$, where B is the gross thickness, w is the net thickness, and $h/2$ is the beam depth. Root radii were 0.001 in. max/in. of gross thickness. The crack starter saw cut, C_0 , is approximately $3B$ and the length L is approximately $13B$. The crack plane of the specimen was normal to the plate surface and parallel to the rolling direction (WR) for all specimens.

Neg 0682600-3

FIGURE 6.1. Double Cantilever Specimen

Test temperatures were maintained on the specimen using two types of cryostats, one with a combination of liquid nitrogen flow control and heaters and a second type using flow control of liquid nitrogen sprayed on the specimen. Thermocouples along the length of the specimens were used to control and monitor temperatures at various crack lengths.

An Instron tensile machine was used to load the specimens. Load and deflection measurements on the specimen were determined from load cell and crosshead displacement measurements of the tensile machine. Crosshead speeds were varied with crack length so that stress intensity rates were held within approximately 10 to 20 $\text{ksi}\sqrt{\text{in.}}/\text{min.}$ A specimen balancing mechanism was used to minimize the nonlinearity in the lower portion of the load extension curve.

The results of the work to date are summarized in Table 6.1 and Figure 6.2 for fracture toughness measurements made on the ASTM A 533 B program plate material. The value $B/(K_{IC}/\sigma_{ys})^2$ in Figure 6.2 is indicative of the ratio between the test specimen thickness and plastic zone size at the crack tip. A significant increase in fracture toughness, K_{IC} , as a function of this ratio can be interpreted as a significant decrease in the plane strain constraint. The value of $B/(K_{IC}/\sigma_{ys})^2$ appears to be about 4 for the lower limit of valid plane strain fracture toughness for A 533 B using the DCB specimen. Since the actual crack runs in the web area of the specimen, a more realistic validity criteria for the DCB specimen thickness would be $w/(K_{IC}/\sigma_{ys})^2$. This would yield a value of 1.4 as a lower limit for $w = 0.35B$, the specimen geometry used in these investigations.

The fracture toughness and arrest temperature dependency are shown in Figure 6.3. The fracture toughness of A 533 B appears to be adequately defined between -160°C and -80°C using the DCB specimen geometry of this study. All data

TABLE 6.1. Fracture Toughness Values of ASTM A 533, Grade B, Class 1 Steel

Specimen Number	Thickness, (B) in.	Temperature, °C	Yield Strength, (b) ksi	K_{Ic} ksi√in.	K_{Ia} ksi√in.	$\frac{B}{(K_{Ic}/\sigma_{ys})^2}$
01KA9 (a)	0.5	-160	117	32.1	29.9	6.64
01KA9 (a)	0.5	-160	117	32.8	30.8	6.36
01KA9 (a)	0.5	-160	117	38.6	29.8	4.59
01KA9 (a)	0.5	-160	117	35.8	32.8	5.34
01KA9 (a)	0.5	-140	105	58.5	31.0	1.60
01KA9 (a)	0.5	-140	105	68.0	(d)	1.19
01KA32 (a)	0.75	-160	117	39.3	28.1	6.64
01KA32 (a)	0.75	-160	117	41.0	33.8	6.14
01KA32 (a)	0.75	-160	117	41.7	30.1	5.90
01KA32 (a)	0.75	-160	117	47.8	28.6	4.48
01KA32 (a)	0.75	-140	105	45.4	31.9	4.18
01KA32 (a)	0.75	-140	105	35.9	32.8	6.40
01KA32 (a)	0.75	-140	105	38.1	55.8	6.00
01KA32 (a)	0.75	-140	105	46.4	(d)	3.84
01KA22 (a)	1.0	-160	117	30.4	29.9	14.84
01KA22 (a)	1.0	-160	117	33.7	31.1	12.02
01KA22 (a)	1.0	-160	117	40.5	35.7	8.36
01KA22 (a)	1.0	-160	117	40.5	37.8	8.36
01KA22 (a)	1.0	-160	117	42.8	36.3	7.46
01KA22 (a)	1.0	-140	105	48.1	37.8	4.75
01KA22 (a)	1.0	-120	97	61.1	56.0	2.52
01KA22 (a)	1.0	-120	97	77.5	(d)	1.57
01KA22 (a) (c)	1.0	-160	117	42.2	33.6	7.70
01KA22 (a) (c)	1.0	-160	117	41.0	39.0	8.16
01KA22 (a) (c)	1.0	-160	117	43.9		8.90
01KA60 (a)	2.0	-160	117	44.0	41.0	14.13
01KA60 (a)	2.0	-160	117	43.6	42.5	14.40
01KA60 (a)	2.0	-140	105	51.3	46.7	8.36
01KA60 (a)	2.0	-140	105	53.7	44.5	7.67
01KA60 (a)	2.0	-120	97	57.3	46.7	5.85
01KA60 (a)	2.0	-100	91	59.8	53.3	4.63
01KA60 (a)	2.0	-80	86	58.5	58.2	4.31
01KA60 (a)	2.0	-80	86	85.7	61.8	2.00
01KA60 (a)	2.0	-60	83	71.5	71.1	2.70
01KA60 (a)	2.0	-60	83	92.8	78.6	1.63
01KA60 (a)	2.0	-40	79	167.8		0.45
01KA20 (e)	1.0	-160	117	37.5	35.1	9.91
01KA20 (e)	1.0	-160	117	44.8	29.5	6.83
01KA20 (e)	1.0	-140	105	41.5	30.8	6.39
01KA20 (e)	1.0	-140	105	41.8	30.4	6.30
01KA20 (e)	1.0	-120	97	39.7	37.5	6.00
01KA20 (e)	1.0	-120	97	55.2	43.0	3.14
01KA18 (e)	1.0	-160	133	43.2	39.2	9.47
01KA18 (e)	1.0	-160	133	59.5	34.3	5.00
01KA18 (e)	1.0	-140	119	57.8	44.4	4.37
01KA18 (e)	1.0	-140	119	66.9	45.7	3.18

(a) Specimen from plate center position in 12 in. A 533 B plate.

(b) Yield Strength values for plate No. 2 of ASTM A 533 B steel.

(c) Specimen was remachined with pin holes and crack starter slot after arm breakage.

(d) Arms of specimen broke.

(e) Specimen 01KA20 from 1/4 T position and 01KA18 from surface of plate.

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FIGURE 6.2. Results of Fracture Toughness Measurements on ASTM A 533 B Plate Material

Neg 0682600-2

FIGURE 6.3. Fracture Toughness Versus Arrest Temperature for A 533 B

plotted are taken from Table 6.1 and are valid within the above established criteria. It is observed that the difference between K_{IC} and K_{Ia} is generally temperature insensitive and not large.

An increase of K_{IC} at the surface of the plate is observed for the data plotted in Figure 6.4. Data for plate center (1/2 T position) is the average of a number of points as shown in Figure 6.3. The invalid point, $B/(K_{IC}/\sigma_{ys})^2 < 4$, at -140 °C was included in the plot since it has the significance that data at -140 °C for 1/4 T and 1/2 T positions from the 1 in. specimen exhibited plane strain validity. The fact that the toughness value was forced to be invalid, even with a higher yield strength at the plate surface, indicates a higher toughness at plate surface.

On the basis of validity criteria established, the DCB specimen offers little size advantage over other types of specimens for use with low yield strength steels. Some qualification is due, since the total number of tests obtained from the DCB specimen would probably be greater for an equivalent volume of material used for other types of specimens. A substantial cost savings could, therefore, be realized using the DCB specimen. Large DCB specimens would probably encounter large fluence gradients over their length during irradiation in most reactors. The fluence gradient might be used to some advantage with DCB specimens as a simple means of measuring the effect of fluence gradients through material properties.

Some additional studies are yet required if the DCB specimen is to be extensively applied to the measurement of fracture toughness of irradiated steels. The specimen might be optimized by increasing the beam height and increasing the net thickness. Increasing the beam height would decrease the maximum fiber stress at the crack plane. Increasing the net thickness would probably increase the temperature range over which valid fracture toughness measurements could be made.

plotted are taken from Table 6.1 and are valid within the above established criteria. It is observed that the difference between K_{Ic} and K_{Ia} is generally temperature insensitive and not large.

An increase of K_{Ic} at the surface of the plate is observed for the data plotted in Figure 6.4. Data for plate center (1/2 T position) is the average of a number of points as shown in Figure 6.3. The invalid point, $B/(K_{Ic}/\sigma_{ys})^2 < 4$, at -140 °C was included in the plot since it has the significance that data at -140 °C for 1/4 T and 1/2 T positions from the 1 in. specimen exhibited plane strain validity. The fact that the toughness value was forced to be invalid, even with a higher yield strength at the plate surface, indicates a higher toughness at plate surface.

On the basis of validity criteria established, the DCB specimen offers little size advantage over other types of specimens for use with low yield strength steels. Some qualification is due, since the total number of tests obtained from the DCB specimen would probably be greater for an equivalent volume of material used for other types of specimens. A substantial cost savings could, therefore, be realized using the DCB specimen. Large DCB specimens would probably encounter large fluence gradients over their length during irradiation in most reactors. The fluence gradient might be used to some advantage with DCB specimens as a simple means of measuring the effect of fluence gradients through material properties.

Some additional studies are yet required if the DCB specimen is to be extensively applied to the measurement of fracture toughness of irradiated steels. The specimen might be optimized by increasing the beam height and increasing the net thickness. Increasing the beam height would decrease the maximum fiber stress at the crack plane. Increasing the net thickness would probably increase the temperature range over which valid fracture toughness measurements could be made.

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FIGURE 6.4. Fracture Toughness Versus Plate Position for A 533 B

Additional work in progress includes an investigation of the effect of round side grooves versus sharp side grooves and the effect fatigue precracking on fracture toughness for DCB specimens.

Irradiation Program

The compact K_{IC} will be used for the determination of irradiation effects on fracture toughness of A 533 B base material during program phase one. The objective of this phase is to determine effects of irradiation at fluences of 1×10^{19} nvt at 450 °F and 5×10^{19} at 450 °F and 550 °F. Compact K_{IC} specimens of the 1T geometry will be used to determine if there are any significant shifts in toughness due to irradiation in temperature ranges where valid measurements can be made.

Initial irradiation capsule designs have been completed for irradiations at both temperatures. The 550 °F irradiations will be conducted in the M-3 hot water loop of the Idaho Nuclear Experimental Test Reactor (ETR). Temperature control will be maintained by gamma heating and control of coolant flow through the capsule. Each specimen temperature will be monitored with thermocouples. Flux monitors will be used to determine the fluence of each specimen, and a dummy specimen will be monitored to determine neutron attenuation through the specimen. The 1T compact K_{IC} specimens are fitted in the capsule with their thickness direction parallel to the axis of the pile and the machined crack opening pointing radially toward the center of the pile. This irradiation orientation will orientate temperature gradients and fluence gradients radially from the subsequent crack line of the specimen, reducing the influence of such gradients along the crack front. The capsule accommodates 30 specimens divided into three subsections of 10 specimens each. Each subsection can be translated or rotated and end to end so that a flux gradients from specimen to specimen can be

minimized. Tubes aligning a flux gradients from specimen to specimen can be minimized. Tubes will hold 30 tensile specimens for use in evaluating mechanical properties at the same conditions of fluence, spectrum, and temperature as fracture specimens.

The 450 °F irradiation will be conducted in a reflector position of the ETR since temperatures this low cannot be obtained in the M-3 loop facility. The irradiation capsules will maintain specimen temperature close to 450 °F with the use of a gas annulus and gamma heating and final temperature control with heaters. The compact K_{IC} specimens, in pairs, will be oriented with their tension testing axis parallel to the axis of the reactor and the machined crack opening facing radially toward the center of the pile. The capsule will accommodate a total of 10 specimens. The temperature of all specimens will be monitored and temperature will be controlled by one thermocouple. Flux monitoring will be similar to that described for the 550 °F irradiation capsule. No fluence gradient at a given crack line will be encountered with the above arrangement of specimens but temperature gradients of a maximum of about 20 °F are expected.

References

1. Heavy-Section Steel Technology Program Semiannual Progress Report for Period Ending August 31, 1967, ORNL 4176. Oak Ridge National Laboratory, Oak Ridge, Tennessee, January, 1967.

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