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NUCLEAR SAFETY QUARTERLY REPORT
FEBRUARY, MARCH, APRIL, 1969
FOR
NUCLEAR SAFETY BRANCH
OF
USAEC DIVISION OF REACTOR
DEVELOPMENT AND TECHNOLOGY

June 1969

AEC RESEARCH &
DEVELOPMENT REPORT

BNWL-1084

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FOREWORD

This report is the ninth 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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BNWL-433	January, February, March	1967
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1. SUMMARY

ENGINEERED SAFETY SYSTEMS STUDIES

Containment Systems Experiment (CSE)

Results of two large scale experiments with containment spray systems are presented. Run A-9 was performed late in the previous quarter to evaluate the effects of very high spray flow rates. Run A-10, performed this quarter, was a demonstration of the effects of continuous spray operation during the fission product release period and for about 27 hr thereafter. In one natural transport experiment, Run A-11, the temperature and pressure of the containment atmosphere were allowed to decay by heat transfer to the environs following fission product release. Results of Run A-11 are not yet available.

Previously reported spray system runs have shown that the removal coefficients for both elemental iodine and particulate materials are directly proportional to spray flow rate during the initial few minutes of spray operation. Run A-9 results confirm this linear dependence up to spray fluxes of about 0.3 gal/min-ft^2 . This spray flux is as high as that being planned for many current power reactor plants. The initial removal half times measured during Run A-9 were 0.57 min for elemental iodine, 1.05 min for cesium, and 2.1 min for uranium. It was observed that the collection efficiency for particles by spray drops decreases with continued spraying, apparently approaching asymptotically a value expected for removal by diffusiophoresis alone.

The effect of diffusiophoresis on particle collection by cold spray droplets was calculated, and the collection efficiency for diffusiophoresis compared with that for diffusion, interception, and impaction for particles of different sizes and for spray drops of 500, 1000, and 2000 μ diameter. The calculation and comparison shows that diffusiophoresis alone

establishes a lower limiting value for particle removal from steam-air atmospheres. For example, for CSE spray run conditions, the removal half life for particles by diffusiophoresis alone is about 30 min. As noted above, observed removal half lives are more rapid than this.

In the continuous spray run A-10, sprays were operated beginning 2 min before fission product release started, and continued for 42 min with fresh, cold spray solution containing boric acid and caustic at pH 9.4. Valving was then changed to recirculate the hot spray solution from the vessel sump through the spray nozzles. Recirculation was continued for 19 hr. At that time, sodium thiosulfate was added to give a 1% concentration; and spraying was continued for 4 hr. Finally, a surfactant material (PEI-1000) was added and a final 4 hr spray performed.

The peak dimensionless concentration for elemental iodine observed at the end of the release period was 0.06, and the average dimensionless concentration over the first 2 hr of the run was 0.024. Both iodine and cesium decreased slowly in gas phase concentration after the end of the release period. The sodium thiosulfate spray produced an 85 min removal half life for methyl iodide compared with the 920 min half life observed during the earlier spray period without thiosulfate. The surfactant was not effective in further increasing the removal rate of methyl iodide.

The pressure reduction effect of sprays was observed during the 42 min cold spray period of Run A-10. The results were compared with predictions of a simple model which assumed thermal equilibrium between the spray drops and the containment atmosphere at all times, and which ignores the heat transferred to the containment atmosphere from the steel vessel walls. The predicted pressure decrease during spraying was about 10% greater than that observed.

During the quarter, a series of coolant blowdown runs was made from the bottom nozzle of the CSE reactor simulator vessel with the simple perforated plate dummy core installed. The runs obtained experimental data for comparison with predictions of the available computer codes used for analysis of coolant blowdown. The results from these runs verified the hypothesis that evolution of dissolved nitrogen was responsible for the extended subcooled blowdown period observed in a run reported last quarter. The predictions of the subcooled decompression regime by the code WATER HAMMER gave reasonable agreement with the experimental results. The code fails to predict some of the observed higher frequency pressure oscillations. These higher frequency oscillations may be evidence of two- or three-dimensional effects; additional pressure measurements are planned to seek confirmation of this possibility. The forces on the dummy core plate are significantly lower than predicted by the WATER HAMMER-SAMIS-RESPON codes during the subcooled decompression. Predictions of the saturated portion of the blowdown by the FLASH2 code give results in reasonable agreement with the experimental results. However, the orifice coefficients used in the code must be modified somewhat for the present two volume configuration from those which were used earlier for the single volume vessel work. The defect indication in the bottom head of the reactor simulator vessel was given an ultrasonic test inspection after each blowdown, and acoustic emission monitoring techniques were used during the current blowdown runs. No indications of defect propagation were obtained.

Particle Transport to Surfaces by Condensing Steam

Hybrid and digital computer solutions were obtained for the previously derived equations describing aerosol transport to a vertical surface in a laminar, condensing steam boundary layer. These solutions permitted particle fluxes to be calculated and compared with predictions from a one-dimensional

approximation. Relative fluxes for different temperature drops are plotted as a function of particle size. Initial effort to evaluate turbulent deposition showed that a diffusion equation including a term for eddy diffusivity would likely express particle fluxes adequately. When eddy diffusivities estimated for a smooth surface were used, deposition rate was lower than in experiments.

PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

Crack Detection in Pressure Piping by Acoustic Emission

Work is in progress to adapt shear wave detection of acoustic emission signals to practical field application requirements. This effort was indicated by previous study of wave propagation characteristics in flat plates. Use of wedge mountings to couple the signal from the plate surface to the transducer enhances signal detection.

Acoustic emission detected during a recent test of carbon steel material showed evidence of a distinction between deformation and fracture generated signals. This correlates with an earlier observation from a pipe burst test conducted at the Battelle-Columbus test site.

A thickness poled, surface mounted sensor with the fundamental resonance at about 1.2 MHz has been fabricated in an effort to enhance the detection of acoustic emission in the low MHz range. Although there is some loss of sensitivity compared to similar lower frequency sensors, initial tests show it to be well within the usable range.

Local application of acoustic emission to investigations of potential slow crack growth by fatigue in process piping is continuing. Three of four joints monitored have shown limited indication of crack activity.

Use of an acoustic emission system to monitor known flaws in the heads of the Containment System Experiment simulator vessel during blowdown tests has continued. Only very limited indications of flaw movement have been detected.

FIXATION OF RADIOACTIVE RESIDUES

During the report period, approximately 4,000,000 Ci of radioactivity were successfully processed in two spray solidification runs in WSEP. Spray Solidification Run SS-8 was made using a simulated Purex Waste 1 (PW-1) feed while SS-9 was made using a simulated Purex Waste 4m (PW-4m) feed. This was the first radioactive demonstration of the PW-4m flowsheet for a relatively clean waste with an exposure of 45,000 MWd/tonne at 30 MW/tonne. Spray solidifier performance during solidification of the new PW-4m flowsheet was excellent.

Simulated PW-4m spray feeds suitable for use in the WSEP were successfully tested in the development spray solidifier. Feed additives of iron, sodium, lithium, and phosphate produced melts gave a volume reduction of eight from solidifying PW-4m at an equivalent 378 liters/tonne. Ruthenium volatilization from low acid PW-4m feed was suppressed by adding solid NaPO_3 flux to calcine in the development melter. The resulting melt in laboratory tests, however, contained a nonmelting rare earth phase which could plug the melter drain valves.

Laboratory tests with PW-4m showed that melting points were lowered as much as 50 °C as the M/P ratio was decreased from 2.75 (the successfully used ratio for both PW-1 and PW-2) to 2.25. The tendency for phase separation in the product was greater at the lower ratio; therefore a compromise of 2.5 was selected for WSEP run SS-9.

Thermal conductivity and dispersibility data for samples of nonradioactive PW-2 phosphate glass show definite improvement for both these characteristics for samples stored at 600 °C as compared to those stored at 100 °C.

2. ENGINEERED SAFETY SYSTEMS STUDIES

CONTAINMENT SYSTEMS EXPERIMENT - G. J. Rogers

Fission Product Transport Studies - R. K. Hilliard

Large-Scale Fission Product Transport Experiments -

L. F. Coleman, R. K. Hilliard, C. E. Linderoth,
J. D. McCormack and A. K. Postma

Two fission product transport experiments, Runs A-10 and A-11, were performed in the large-scale CSE containment vessel during this report period. The first was a containment spray test in which the sprays were operated continuously for 24 hr, beginning just before fission product simulants were released. In the second test no sprays operated, steam feed was stopped immediately after fission product simulant release was completed, and the temperature and pressure were allowed to decrease by natural heat transfer. In addition, results of Run A-9 (a containment spray experiment made during the previous quarter) were evaluated. Runs A-9 and A-10 are discussed in the following paragraphs. Run A-11 will be reported next quarter.

Run A-9. The primary purpose of Run A-9 was to demonstrate the removal of iodine and aerosol particles from containment atmospheres at a high spray flow rate. Theory predicts that removal rate constants for both gases and particles should be directly proportional to spray flow rate, unless some other transfer parameter is altered by increased flow. One possible alteration would be change of spray drop size distribution by coalescence of drops at high spray fluxes. Parsly⁽¹⁾ has concluded that less than a 20% error would be introduced by neglecting coalescence at spray fluxes up to about 0.15 gal/min-ft², but experimental verification is needed. The CSE spray experiments substantiate this conclusion for fluxes up to 0.3 gal/min-ft².

The experimental conditions of CSE spray tests A-3 through A-9 were reported earlier.⁽²⁾ Three flow rates to the 25 ft diameter vessel were used: 12.8, 49, and 145 gal/min. In Run A-9 the highest flow was used, corresponding to a spray flux of 0.30 gal/min-ft². This is about the rate for many currently proposed nuclear power reactor containment systems. The initial atmosphere was a saturated steam-air mixture at 250 °F, 44 psia. Fresh room temperature water with boric acid and caustic, pH 9.4, was used. Twelve Spraying Systems Co. spray nozzles No. 3/4 A50 (hollow cone) were used at 50 psi differential, giving a mass median diameter of 1220 μ . The iodine, methyl iodide, cesium, and uranium oxide were released into the vessel and allowed to mix for 30 min. The spray system was operated for 3 min. A second 3 min spray and then a 10 min spray was performed for a total of 2220 gal of fresh spray solution. Finally, the spray liquid in the sump was recirculated for one hour.

Figures 2.1, 2.2, 2.3, and 2.4 show the time history of concentrations in the gas phase for elemental iodine, methyl iodide, cesium, and uranium, respectively. The concentration half lives during non-spray periods were determined by the best fit of the experimental measurements, and the half lives during spray periods were calculated as described previously.⁽²⁾ Elemental iodine was removed very rapidly during the first short spray period ($t_{1/2} = 0.57$ min); very little further reduction in gas phase concentration was effected by subsequent sprays. Methyl iodide was not removed significantly during the three fresh spray periods. The reduction in concentration in the main room is accounted for by diffusion into the lower rooms. Cesium was removed rapidly during the first spray ($t_{1/2} = 1.05$ min) with less rapid but still significant removal during subsequent sprays. Uranium was removed with a 2.1 min

FIGURE 2.1. Elemental Iodine Gas Phase Concentration, Run A-9

FIGURE 2.2. Methyl Iodide Gas Phase Concentration, Run A-9

FIGURE 2.3. Cesium Gas Phase Concentration, Run A-9

FIGURE 2.4. Uranium Gas Phase Concentration, Run A-9

half life during the first spray. Samples for subsequent spray periods have not yet been analyzed for uranium at the time this report was written.

The effect of spray flow rate on λ_s , the washout coefficient, is shown in Figures 2.5, 2.6, and 2.7 for elemental iodine, cesium, and uranium, respectively. Experimental values for λ_s , determined for the first spray periods of five CSE experiments, are plotted against flow rate. The curves represent the theoretical dependence on flow rate, calculated from Equation (1) for elemental iodine absorption and from Equation (2) for particle removal.

$$\lambda_s = \frac{6 F h_{avg} k_c}{V U_t d_{SMD}} , \quad (1)$$

where

- F = spray flow rate to vessel, ft^3/min
- h_{avg} = average fall height of spray drops, ft
- k_c = gas phase mass transfer coefficient, ft/min
- V = volume of contained gas, ft^3
- U_t = terminal settling velocity of mean spray drop, ft/min
- d_{SMD} = surface mean diameter of spray drops, ft

$$\lambda_s = \frac{3 F h E}{2 V d_{SMD}} , \quad (2)$$

where E = drop collection efficiency, dimensionless.

Two curves are plotted on Figure 2.5 because two atmospheric conditions were used: Runs A-3 and A-4 with room temperature air and the others with a steam-air mixture initially at 250 °F and approximately 48 psia. The washout coefficient is linear with flow rate, in agreement with theory.

Figures 2.6 and 2.7 show similar data for the particulate materials cesium and uranium. The washout coefficient for

FIGURE 2.5. Effect of Spray Flow Rate on Washout Coefficient for Elemental Iodine

FIGURE 2.6. Effect of Spray Flow Rate on Washout Coefficient for Cesium

FIGURE 2.7. Effect of Spray Flow Rate on Washout Coefficient for Uranium

particles is dependent on E , the drop collection efficiency, a factor which usually is not known precisely. Therefore, two theoretical curves are shown on each figure for two values of efficiency which span the observed results. Prediction of efficiencies is discussed in a later section of this report. The washout coefficients observed during the first spray periods are seen to be essentially linear with spray flow rate.

The rapid washout of particles observed during the first spray periods was not maintained during subsequent sprays. In each succeeding spray period the washout coefficient was smaller than in earlier sprays. This behavior is shown for cesium in Figure 2.8, where the drop collection efficiency is plotted against accumulated volume of liquid sprayed for all comparable CSE tests. The drop collection efficiency decreased from a high value of about 0.06 to 0.01 at the end of spraying. It is interesting to note that the curve representing the average of experimental results appears to be approaching an asymptotic value of 0.005 at long spraying times, which is the value predicted for diffusiophoresis alone.

Run A-10. In all CSE spray tests through Run A-9, the fission product aerosol simulant was released first and allowed to mix and be transported by natural processes for 30 or 40 min before the spray was started. The sprays were then operated intermittently for short times. This method of operation was best suited for investigation of the effect of transport parameters, but is not the way that containment spray systems are intended to be operated in the event of an accident.

The purpose of Run A-10 was to demonstrate the performance of a containment spray system for removing iodine and particles when the spray is operated continuously, starting before the release of fission product materials.

FIGURE 2.8. Drop Collection Efficiency as a Function of Liquid Volume Sprayed

The spray system and nozzle arrangement was the same as used in Runs A-4, A-6, and A-7 (twelve Spraying Systems Co. 3/4 7G3 nozzles operating at 40 psia). Fresh, 80 °F spray solution of boric acid and sodium hydroxide, pH 9.4, was started 2 min before iodine and cesium release began, and continued for a total of 42 min, delivering a spray volume of 2100 gal. The fresh spray solution injection was ended, and valves were set for recirculation of solution from the sump. Recirculation spray started 10 min later and continued for a total of 19 hr. At this time, enough sodium thiosulfate was added to the sump to give a 1 wt% $\text{Na}_2\text{S}_2\text{O}_3$ solution and recirculation was continued for 4 more hours. A surfactant material (Dow Corning PEI-1000) was then added to the sump solution and recirculated for four more hours.

Figure 2.9 shows that the elemental iodine concentration rose to a maximum of about 10 mg/m^3 at the end of the release period, and then it decreased slowly (220 min half life) for the duration of the experiment. Since 95 g of elemental iodine was injected into the 595 m^3 main room of the containment vessel, the maximum dimensionless concentration was 0.06. The dimensionless concentration C/C_0 averaged for the first 2 hr period was 0.024; the 24 hr average was 0.006. Cesium behavior closely paralleled that of iodine.

Methyl iodide was removed from the atmosphere at a slow, steady rate, the half life being 920 min during the 19 hr recirculation with the boron-caustic spray solution. Figure 2.10 shows that adding sodium thiosulfate caused the concentration of methyl iodide to decrease much faster, the half life during the 4 hr period being 85 min. Adding the surfactant PEI-1000 did not increase the removal rate, since the half life was 155 min with the surfactant added.

FIGURE 2.9. Elemental Iodine Gas Phase Concentration, Run A-10

FIGURE 2.10. Methyl Iodide Removal by Special Sprays, Run A-10

Effect of Diffusiophoresis on Particle Collection by Cold Sprays - A. K. Postma and J. D. McCormack

Diffusiophoretic motion of aerosol particles is potentially an important mechanism of particle collection for cold water sprays. Although precise prediction of the effect of steam condensation on a particle is not easily accomplished, a simple model may be used to estimate the effect of steam condensation on drop collection efficiency.

At the surface of a drop, the deposition rate due to diffusiophoresis is

$$\text{deposition rate} = 4\pi r_d^2 C_A V_D , \quad (3)$$

where r_d = drop radius

C_A = particle concentration at drop surface

V_D = diffusiophoretic deposition velocity.

Integrating Equation (3) to get the total amount of aerosol deposited gives

$$\text{aerosol deposited} = 4\pi r_d^2 C_A \int_0^t V_D dt . \quad (4)$$

In carrying out the integration, we have assumed the aerosol concentration to be constant during the drop existence time. The diffusiophoretic velocity for the slip flow regime is given by Waldmann and Schmidt⁽³⁾ as

$$V_D = (1 + \sigma_{12} \gamma_2) \frac{D}{\gamma_2} \frac{d}{dx} (\gamma_1) \Big|_{\infty} , \quad (5)$$

where $\sigma_{12} = 0.95 \frac{m_1 - m_2}{m_1 + m_2} - 1.05 \frac{d_1 - d_2}{d_1 + d_2}$

γ_2 = mole fraction of nondiffusing species

γ_1 = mole fraction of diffusing species

D = diffusivity of gas mixture

$\left. \frac{d}{dx} (\gamma_1) \right|_{\infty}$ = gradient in diffusing species at points far from the particle

m_1 = molecular mass of species 1

m_2 = molecular mass of species 2

d_1 = molecular diameter of species 1

d_2 = molecular diameter of species 2.

The rate of vapor condensation at the drop surface is

$$N_S = \frac{4\pi r_d^2 \rho D}{\gamma_2} \left. \frac{d\gamma_1}{dr} \right|_{r=r_d} \quad (6)$$

where

N_S = steam transfer rate to drop

ρ = total gas concentration, moles/cm³.

The total amount of water condensed on a drop may be obtained by integrating Equation (6) with respect to time:

$$\text{amount of water condensed} = 4\pi r_d^2 \rho \int_0^t \frac{D}{\gamma_2} \left. \frac{d\gamma_1}{dr} \right|_{r=r_d} dt. \quad (7)$$

If it is now assumed that the particles do not influence the vapor concentration gradient, then

$$\left. \frac{d\gamma_1}{dr} \right|_{r=r_d} = \left. \frac{d}{dx} (\gamma_1) \right|_{\infty}, \quad (8)$$

If, in addition, one assumes that the drops increase in temperature due only to steam condensation, by an amount ΔT , the total integrated steam flux may be related to ΔT by a heat balance. Use of the equations above gives for the effective collection efficiency,

$$E = \frac{4(1 + \sigma_{12} \gamma_2) \rho_\ell r_d C_{p\ell} \Delta T_\ell}{3 \Delta H_v \rho h} \quad (9)$$

where E = drop collection efficiency

ρ_l = liquid density

Cp_l = liquid heat capacity

ΔT_l = temperature rise experienced by a drop

h = drop fall height

ρ = total gas density, moles per unit volume

ΔH_v = latent heat of condensation of vapor.

Equation (9) was evaluated numerically for conditions encountered in CSE:

$$\rho_l = 0.94 \text{ g/cm}^3$$

$$Cp_l = 1.01 \text{ cal/g } ^\circ\text{K}$$

$$\Delta H_v = 9470 \text{ cal/mole}$$

$$\rho = 1.03 \times 10^{-4} \text{ moles/cm}^3$$

$$\Delta T_l = 83 \text{ } ^\circ\text{C}$$

$$h = 1030 \text{ cm}$$

$$r_d = 250 \text{ } \mu, 500 \text{ } \mu, 1000 \text{ } \mu.$$

The results of this calculation are presented in Figure 2.11, where drop collection efficiency is shown as a function of particle size.

The drop collection efficiencies for diffusion, interception, and impaction are shown for comparison. The diffusional deposition was calculated from the Frössling equation, as modified by Ranz and Marshall.⁽⁴⁾ Interception was calculated on the basis of potential flow,⁽⁵⁾ and impaction from the results of Ranz and Wong.⁽⁵⁾

The significant feature shown by Figure 2.11 is that diffusiophoresis is an important mechanism for a wide range of intermediate particle sizes. Differences in collection efficiency for the three drop sizes disappear if the washout half-time, where $t_{1/2} = (0.693 \text{ dV}) / [(3/2) \text{ FhE}]$, is calculated. Thus, the diffusiophoretic effect will establish a minimum washout rate, independent of drop size, particle size, and fall height.

FIGURE 2.11. Aerosol Washout Calculated for Cold Water Sprays Entering a Containment Atmosphere Saturated at 250 °F

For the CSE vessel, with a spray flow rate of 50 gal/min, and an inlet liquid temperature of 100 °F, the washout rate for diffusiophoresis is predicted to be about 30 min, a significant removal rate.

It is interesting to note that for cold sprays, washout rates measured in the CSE vessel are faster than predicted for diffusiophoresis alone. Thus, it appears that other mechanisms must be operating to cause particle removal by cold sprays. One explanation would be that the particles exist in fog drops which are large enough to be collected by inertial impaction. Another possibility is that the several operating mechanisms combine to give an overall washout rate greater than the sum predicted for the individual mechanisms.

In the CSE experiments the steam content of the atmosphere has been nearly constant; hence the diffusiophoresis rate will remain important as long as ΔT is maintained. In a system where the steam is depleted from the atmosphere the diffusiophoresis rate will decrease until the diffusion or interception efficiencies determine the minimum removal rate.

Pressure Suppression by Water Sprays in the CSE Vessel -

A. K. Postma

The primary purpose of water sprays provided as part of many reactor containment systems is to condense steam involved during a loss-of-coolant accident. Steam condensation may be required to prevent pressurization of a containment vessel beyond its design limit. Because relatively little information is available to demonstrate the thermal response of a large building atmosphere to a cold water spray, one of the aims of the CSE studies is to provide experimental information applicable to the evaluation of large scale containment systems.

Run A-10 of the CSE aerosol transport series provided an opportunity to study the quenching of steam within the CSE

vessel by a cold water spray of appreciable duration. In this run, water at about 80 °F was sprayed into the vessel at a rate of 50 gal/min for a time period of 42 min. Initial atmosphere temperature was 249 °F. Steam was added to the vessel at a rate of 360 lb/hr during the first 10 min of the spray period, and was then increased to 690 lb/hr during the remainder of the spray period.

The average temperature of the containment vessel atmosphere during the spray period is shown in Figure 2.12 as a function of time. During the spray period, temperatures at all locations within the main room (including an unsprayed region above the spray header), the drywell (lid open), and the middle room were within several degrees fahrenheit of the average temperature. Thus, the atmosphere within these regions was well mixed. The temperature within the lower room was at 171 °F at the beginning of the spray period and increased to 190 °F at the end of the spray period.

The efficiency of a water spray for cooling an air-steam mixture depends on the degree to which the spray drops achieve thermal equilibrium with the containment atmosphere during their fall. Experimental measurements designed to determine the drop temperature as a function of fall height were carried out during Run A-10. The method used was to collect spray liquid in a double-walled pyrex flask, insulated to inhibit heat transfer to collected drops. The temperature of the collected water was measured by a thermocouple inserted into the U-tube outlet of the collector flask. The liquid volume held up within the collector was about 5 ml, from which it was estimated that a temperature lag of about 1 min would be caused by mixing within the collector.

Drop collectors were installed at elevations corresponding to vertical distances from the spray nozzles of 4, 16, and 28 ft.

FIGURE 2.12. Temperature Decay due to Cold Water Spray in CSE Vessel

The measured temperatures within the collected liquid were higher at the three locations than the atmosphere temperature as measured by thermocouples shielded from the spray by sampler cluster housings. The temperature measured within the drop collectors was consistent with behavior expected if the drops were in thermal equilibrium with the vessel atmosphere at the three vertical positions.

Heat transfer calculations based on limiting case models (stagnant drops, and perfectly mixed drops) predict very rapid heat-up for small drops. For drops as large as 2000 μ diameter, however, the calculations indicate that relatively large fall heights would be required to attain thermal equilibrium.

The pressure suppression efficiency of the water sprays was evaluated in a preliminary way by comparing the measured pressure response with that calculated for a naively simple model. In this model, the spray water is assumed to be in thermal equilibrium at all times with the vessel atmosphere. Also, heat transfer from the vessel walls is ignored. The measured pressure is compared to the pressure predicted by means of the simple model in Figure 2.13.

The predicted pressure decrease is about 10% greater than the observed pressure decrease. This discrepancy is no doubt due mostly to heat transfer into the atmosphere from the steel walls of the vessel. A more refined mathematical analysis, accounting for transient heat conduction through the vessel wall and external insulation, is underway.

FIGURE 2.13. Pressure Decay due to Cold Water Spray in CSE Vessel

Small-Scale Fission Product Transport Experiments -

R. H. Bond

A mathematical model for predicting the rate of removal of elemental iodine from containment atmospheres was described previously.^(6,7) One assumption in the model is that elemental iodine is transferred to all surfaces exposed to the steam-air-iodine atmosphere during the initial period when gas phase diffusion across boundary layers is the rate-limiting step. Large containment vessels have 2 to 4 times more total surface area than that of their exterior, geometrical surfaces. Thus, this assumption results in predictions of removal rates several times greater than if only the external surface area is used. Tests in the CSE (Runs A-1, A-2, and A-5) have substantiated the validity of this assumption, but the total surface area of the CSE vessel is only 50% greater than its external surface. Additional experimental evidence is considered desirable.

An effort was made to demonstrate that noncondensing surfaces are equally important as condensing surfaces for diffusion. Tests were made in the Aerosol Development Facility SAT vessel, first with all surfaces condensing and then with approximately 50% added stainless steel surface in the form of thin sheets hung in the vapor space and on which no steam condensed. Unfortunately, for the primary purpose of these tests, most of the iodine was found to be associated with particles, and the elemental iodine behavior was masked by the more abundant particulate aerosol. Tests are being repeated with the cesium and uranium-Zircaloy particles omitted so that elemental iodine behavior can be studied without interaction with particles.

In these tests the gas concentration half lives of particulate aerosols (cesium, uranium, and particulate iodine) were the same with and without the extra noncondensing surfaces. The conclusion can be made that the mechanisms responsible for

removing the particles from the gas phase are not dependent on the presence of vertical, noncondensing surface. This was predictable for gravitational settling and diffusiophoresis, but is new information concerning turbulent diffusion because it shows the latter to be insignificant for these test conditions. It does not exclude the possibility that turbulent diffusion may be significant to the main vessel walls, where convection velocities should be greater and thus boundary layer thickness should be smaller.

Coolant Blowdown Studies - R. T. Allemann

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

During this report period a series of blowdown runs was made from the bottom nozzle with a perforated plate dummy core installed in the vessel. The purpose of the series is to study the effects on the subcooled and saturated phases of coolant blowdown of a simple perforated plate separator representative of mixer, support, and grid plates used in nuclear reactors.

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

Blowdowns were made from the bottom nozzle of the CSE simulator vessel with an elbow, extension, and orifice plate

as shown in Figure 2.14. The core plate divided the vessel volume to give a ratio of upper to lower volume of 2.3. Sixty four nozzles of 1.23 in.² each were installed with the remaining 24 holes plugged as shown in Figure 2.15. The flow area of this sieve plate was 0.55 ft². Blowdown was initiated using the double-rupture-disk, over-pressurization technique. The experimental arrangements for the blowdown series reported here is described extensively in the previous quarterly report.⁽²⁾

Pressure and temperature sensors are located in nozzles J, H, R (refer to Figure 2.14), the instrument ring, and in taps on the nozzle extension piece. Additional thermocouples are located along the length of the liquid level probe and are designated TL1, TL3 to TL6 in the figure. A special thermocouple assembly is installed in nozzle "O" for transient heat transfer measurements. Four of the eight support lugs for the dummy core plate are instrumented with strain bolt assemblies for measuring the integrated force on the plate. The center of the core plate and one of the lugs also have strain gages installed. Additional strain gages are installed on the inside and outside of the area about the base of nozzle C. Each of these strain gages has a thermocouple attached to the vessel skin in its vicinity. A differential pressure sensor is attached to tubing routed through the instrument ring to one of the orifices inside the vessel. A second differential pressure sensor is attached to pressure taps 10 in. apart in the center of the nozzle extension piece. A linear displacement device is installed on the bottom of the nozzle extension piece to monitor the motion of the nozzle during blowdown. In addition to these sensors, acoustic emission transducers are installed on the top and bottom vessel head to monitor any defect propagation in the bottom head.

The initial conditions for the blowdown experiments performed this period are summarized in Table 2.1.

FIGURE 2.14. CSE Reactor Simulator Vessel with Core Plate

64 SYMMETRICALLY LOCATED ORIFICES AS
MARKED "O" ON ONE-FOURTH OF THE SKETCH

44 SYMMETRICALLY LOCATED PLUGS AS CROSS-
HATCHED AND MARKED "P" ON ONE-FOURTH OF
THE SKETCH

FIGURE 2.15. Sieve Plate Plug and Nozzle Locations

TABLE 2.1. Blowdown Run Conditions

Run No.	Nominal Break Size, in.	Break Area, ft ²	Initial Pressure, psia	Initial Temp., °F	Gas or Steam Volume, ft ³
B-19B	2	0.0156	2115	603	0
B-20B	4	0.0643	968	500	0
B-21	6	0.146	990	500	0
B-24	4	0.0643	809	545	50

The purpose of these runs was to obtain data for comparison with predictions of computer codes used for analysis of coolant blowdown. Of particular interest was data on the force on the perforated plate dummy core. In addition, the defect indication in the bottom head of the reactor simulator vessel was monitored for evidence of propagation.

Run B-19B was a repetition of Run B-19A reported last quarter.⁽²⁾ The extended time (about 25 sec) to reach saturation pressure in Run B-19A was suspected to have resulted from nitrogen dissolved in the water. Therefore, the operating procedures for Run B-19B were revised to eliminate the dissolved nitrogen. Results for Run B-19B, discussed in a later section, confirmed that dissolved nitrogen was responsible for the slow subcooled decompression. Accordingly, the revised operating technique was adopted for all runs starting with a full vessel.

Because of the uncertain adequacy of the mathematical models and computer codes used to predict the transient differential pressures and the resultant forces on the dummy core plate, the break size and initial pressures were limited to provide substantial margins against possible overloading of the core support lugs in the reactor simulator vessel. For example, when the plate force was assumed to be proportional to the initial pressure and break area, an estimate was made for the maximum load of Run B-20 based on the peak load measured for Run B-19B.

Since the maximum load measured for B-19B was 18,850 lb, the estimated maximum load applied to the plate for B-20B is 36,340 lb, which should be acceptable. The plate support lugs are designed to support a load of 100,000 lb. In similar fashion, the peak measured forces on the core plate from B-19B and B-20 were used in estimating the forces expected in Run B-21 with a 6 in. nominal break size.

Data and results of analyses for the run series are shown in Figures 2.16 through 2.23. The fluid state data in Figures 2.16, 2.17, 2.18, 2.19, and 2.20 show that the fluid in the vessel stays close to saturation after the initial decompression. The orifice pressure data in Figures 2.21, 2.22, and 2.23 indicate the rather complex pressure patterns that occur immediately after the rupture disk breaks. The "break time" used for decompression analysis code input is taken from this leak orifice pressure history. It is seen that a simple ramp-function area increase break is perhaps overly simplified. Note that the pressure dips well below saturation before beginning to recover toward its exit critical value.

Vessel Pressure. The FLASH2 code⁽⁸⁾ has been applied to four CSE blowdown Runs B-19B, B-20B, B-21, and B-24. A two-volume approximation was used with previously obtained leak-orifice coefficients of 0.82 for Run B-19B, 0.69 for Runs B-20B and B-24, and 0.57 for Run B-21. The orifice coefficient for Run B-21, a 6 in. nominal break (5.187 in. diameter), was predicted within 3% of the correct coefficient by the CSE correlation⁽⁹⁾ developed from single-volume bottom blowdown data.

The predicted pressure decay curves agreed relatively well with the experimentally measured curves (Figures 2.24, 2.25, 2.26, and 2.27). The B-21 prediction was slightly above the experimental curve in the two-phase blowdown region. This indicates that the initial temperature used for the prediction was

FIGURE 2.16. Fluid Temperature History B-19B

FIGURE 2.17. Fluid State During B-19B

PRESSURE AT NOZZLE R, PSIG (P-6)

FIGURE 2.18. Fluid State B-20B

FIGURE 2.19. Fluid State B-21

FIGURE 2.20. Fluid State B-24

FIGURE 2.21. Pressure at Orifice B-19B

FIGURE 2.22. Pressure at Orifice B-21

FIGURE 2.23. Pressure at Orifice B-24

FIGURE 2.24. Vessel Pressure History B-19B

FIGURE 2.25. Vessel Pressure History B-20B

FIGURE 2.26. Vessel Pressure History B-21

FIGURE 2.27. Vessel Pressure History B-24

too high. All the predictions showed some discrepancy between the measured and predicted time of transition to single-phase saturated flow from two-phase flow. Table 2.2 contains a list of predicted and measured times to the transition for full-vessel bottom blowdowns with a sieve plate for Runs B-19B, B-20B, and B-21. Estimated correct coefficients based on the transition times are also listed. These corrected coefficients have been fitted with the following empirical expression:

$$C_{M2} = 0.325 R^{-0.142}$$

C_{M2} = flow coefficient applied to the Moody model⁽¹⁰⁾ of mass velocity based on CSE, two-volume data.

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

Results from the B-24 prediction were not used because there was some uncertainty in the initial liquid volume. The input value for initial liquid volume would directly affect the predicted transition time. Any comparison between predicted and measured transition time for B-24 is therefore questionable.

For most of the pressure predictions, the slope of the decay curve in the saturated single-phase (high quality) region was too shallow, particularly in the range of 20 to 300 psig. This anomaly was probably caused by using a two-phase orifice coefficient, which was determined by adjusting transition times for previous predictions,⁽²⁾ over the entire duration of the transient. The problem could be corrected by modifying the code to allow use of two coefficients, one for two-phase flow and one for saturated single-phase flow.

Two FLASH2 trials were made for a B-24 prediction. Experimentally determined initial conditions for the run were 490 °F, approximately 103 ft³ initial liquid volume, and 825 psia (including about 120 psi nitrogen overpressure). Using the same initial pressure with the upper volume partially filled

TABLE 2.2. Two-Phase Saturated Critical Flow Orifice Coefficients

Run No.	Break Diameter, in.	Transition Time, sec		C_M Orifice FLASH2 (a)	C_{M2} Coefficients Corrected (b)	Time When Liquid Level Reaches Plate, sec	
		Experimental	FLASH2			Experimental	FLASH2
B-19B	1.689	59.5	58.0	0.82	0.800	41	54
B-20B	3.438	26.5	25.5	0.69	0.664	24	26
B-21	5.187	13.5	13.8	0.57	0.583	11.8	13.8
B-24	3.438	17.5	17.0	0.69	-----	14	16

a. Best C_d from previous comparisons of blowdowns with no sieve plate.

b. Estimated C_d based on corrected transition times for blowdowns with sieve plate.

(saturated conditions) and the lower volume at 490 °F for the FLASH2 prediction gave poor results based on the comparisons of predicted and measured pressure histories. Further analysis of the data showed that the vessel pressure dropped to nearly saturation (740 psia) very rapidly. Therefore, a second prediction was made using 740 psia initial pressure. The results agreed well with the experimental data. However, the inability of FLASH2 to handle a case with nitrogen overpressure was accentuated. This fact coupled with the known inability of FLASH2 to accurately predict the results of a subcooled, full-vessel transient with dissolved nitrogen (B-19A)⁽²⁾, and the possible use of dissolved gases to lower transient forces, point to the need for modifications to FLASH2 to properly handle these cases. These modifications are currently being studied.

Mass Flow Rate. The initial depressurization impulse sets the entire vessel into vertical oscillation which continues throughout a blowdown. The oscillation is the dominant feature of the weight load cell output and must be eliminated by filtering and hand smoothing to obtain a curve of the weight of water left in the vessel versus time.

Because of the large fluctuation in the experimental data, it is difficult to compare accurately the predicted total weight in the vessel with the measured data. But, considering this source of uncertainty, the agreement between FLASH2 predictions and observed results seems satisfactory, as shown in Figure 2.28. Again, the poorest agreement was at the times near and after transition. If the modified orifice coefficients had been used, better weight agreement would have been obtained. However, a second orifice coefficient would still be required for the high quality post transition regime. Figure 2.29 shows a consolidation of mass flux values, for the saturated regime, determined by taking the slope of the weight data. Time is dimensionalized to correct for differences in initial mass, flow area, and

FIGURE 2.28. Reduced Mass of Liquid Remaining in Vessel

FIGURE 2.29. Mass Flux History

discharge coefficient. The system does not have sufficient frequency response to obtain subcooled flow rates. The times of transition to steam flow are indicated by the sharp decrease in mass flux. These times agree with the time of the pressure history slope inflection.

The lower flow rate for large area breaks may be explained on the basis that higher total flow rates sweep more bubbles into the nozzle, reducing the specific weight and hence the mass flux. The pressurization of the vessel by nitrogen at the beginning of Run B-24 allowed not only the free nitrogen but also dissolved nitrogen to maintain an overpressure for the entire duration of the blowdown (See Figure 2.20).

The transition is not too evident on B-21 weight data because of the very small fl/D ratio. With this small ratio the mass flux of the single-phase steam is approximately equivalent to the mass flux of the high quality two-phase fluid.

Liquid Level. The liquid level prediction did not agree with the time domain reflectometer (TDR) measured level histories for any of the runs as illustrated in Figures 2.30 and 2.31. Table 2.2 contains a list of observed and predicted times when the level dropped through the sieve plate. In all cases, the predicted time was too long. For two full-vessel blowdowns, B-19B and B-20B, the transition time was greater than predicted, while the time when the level dropped through the plate was less than predicted. Therefore, the problems with the level predictions were probably caused by the use of too high a resistance for the core plate. The increase in liquid level shown by the data from the TDR probe (Figure 2.30) is not real. This is believed to be a result of the rapid temperature change in the vessel after transition. This rise should occur only after transition, but in Run B-20B it anomalously began immediately after the liquid level dropped through

FIGURE 2.30. Normalized Liquid Level History

FIGURE 2.31. Normalized Flash-2 Predictions

the plate at about 25 sec. It appears from the pressure data that although true transition had not begun at 25 sec since the average pressure was still decreasing slowly (Figure 2.25), the outlet nozzle pressure at 25 sec was experiencing some large pressure fluctuation, perhaps indicative of incipient transition in the lower chamber while liquid is still present in the upper chamber. This liquid, in being swept toward the bottom side exit, did not show up as an indication on the TDR which responded only to the temperature changes then occurring.

Nitrogen Solubility Effects. Two techniques have been used to pressurize the water in the reactor simulator vessel above the saturation condition.

The first technique was to partly fill the vessel with cold water, and pressurize the system with nitrogen through pipes on the top of the vessel. During the course of heating, the water which was being circulated through a high temperature heat exchanger expanded to fill the vessel, thus displacing all the nitrogen. Minor adjustments in final level were made by bleeding or injecting water. The high-pressure nitrogen was in direct contact with a large surface area of circulating heated water for several hours, and thus had ample opportunity to dissolve in the water.

The second technique was to fill the vessel all the way into the accumulator (Figure 2.14) with cold water and pressurize with nitrogen. All water expansion volume caused by heating was allowed to bleed off to maintain the water-level in the accumulator. A heat exchanger, used to keep the water (which is in contact with the nitrogen) cold, and about 12 ft of 0.24 in. ID tubing isolated the nitrogen from the main vessel water.

Figure 2.32 shows that the solubility of nitrogen goes up with temperature, but that pressure is still the main parameter which determines the amount dissolved.

FIGURE 2.32. Solubility of Nitrogen in Water at Varying Temperatures

Figures 2.33 and 2.34 present experimental results showing the effect of an operating technique in which nitrogen could dissolve in the water versus one in which nitrogen dissolution was kept minimal.

Figure 2.33 shows the pressure just below the perforated plate in the main vessel during the initial part of the sub-cooled decompression for two similar blowdowns except for the technique used in pressurizing.

The nitrogen (assumed to be dissolved in the water) slows down the rate of decompression to saturation (30 sec versus 0.5 sec). It reduces the magnitude of the high frequency pressure oscillation by a factor of four (48 psi versus 12 psi), and the size of the lower frequency pressure drop steps about 50 psi to 25 psi at the beginning of the decompression.

Figure 2.34 shows (for the two blowdowns) the high speed output values of strain (which is proportional to force applied) obtained from a strain gage mounted on a bolt used to support the perforated plate in the reactor vessel simulator. The nitrogen apparently reduced the effective force on the perforated plate by a factor of two or more. The sharp steps are caused by digitizing very low sensor output. Occasional spikes are believed to be switching transients, not real strain values.

Other results obtained from these blowdown tests show that nitrogen has an effect on the rate of liquid level decrease in the vessel and that its effect on the total blowdown time is small.

WATER HAMMER predictions shown on Figure 2.33 give good agreement for the general decompression rate when no nitrogen is present.

Forces on the Core Plate. Strain bolts supporting the core plate are used to estimate the average differential pressure on the core plate during the transient decompression. The

FIGURE 2.33. Pressure in Vessel During Subcooled Decompression

FIGURE 2.34. Strain in Bolt Supporting the Perforated Plate in the CSE Reactor Simulator Vessel

signal outputs of these bolts were calibrated before the run series by applying known loads to the plate and relating these loads to the equivalent pressure on the surface of the plate. Estimates of pressure on the plate by this method are compared to predictions of the code WATER HAMMER. Two other computer codes are involved in the analysis of the plate loading. The first (SAMIS)⁽¹¹⁾ is used to determine natural frequencies, mode shapes, and associated stresses of the perforated plate and support bolt system. The second (RESPON) is used to get the response of the plate and bolts to a driving function such as the output of the code WATER HAMMER. This program solves the uncoupled modal response equations of the plate system subjected to an arbitrary load. A consolidation of the reduced strain bolt outputs, WATER HAMMER pressure predictions, and RESPON support bolt stresses appear in Table 2.3. The table is constructed to show the nominal peak observed and predicted values and the approximate ratio of the predicted to observed values. A sample output of the WATER HAMMER pressure loading and a strain gage loading for B-21 are shown in Figure 2.35. The wave forms of the strain gages agree well with each other in a given run. However, the magnitude of the peak values predicted are 3 to 5 times the observed values (Table 2.3), and the wave form of the load history and the predicted history are somewhat dissimilar. Outputs of the code RESPON also tend to overestimate the bolt loading by factors of 5 to 12. The preliminary explanation for this result is that the driving function in the real case is not as severe as that in the code. Further analysis is required to determine the influence of three dimensional effects in the bottom blowdown case and the degree of pressure wave interference and attenuation that occur in the process of plate loading.

TABLE 2.3. Consolidation of Plate Loading Information

Blowdown Number and Conditions	Strain Gage	(1) Observed Peak Differential Pressure From Bolts	(2) WATER HAMMER Peak Differential Pressure, psi	Ratio (2/1)	(3) Observed Bolt Stress, psi	(4) RESPON Predicted Bolt Stress, psi	Ratio (4/3)
B-19B	SB5I	17.1	52.	3.04	2860	15000	5.2
2 in.	SB6I	18.5	52.	2.81	2680	15000	5.6
2135 psia	SB8I	12.0	52.	4.33	3830	15000	3.9
600 °F	SB8Ø	10.4	52.	5.0	3890	15000	3.9
		$\overline{\Delta P} = 14.5$					
B-20B	SB5I	17.8	43.	2.42	4080	46000	11.3
4 in.	SB8I	16.1	43.	2.67	5130	46000	9.0
983 psia	SB8Ø	9.9	43.	4.34	3680	46000	12.5
500 °F		$\overline{\Delta P} = 14.6$					
B-21	SB8I	43.2	75.	1.74	9240	99000	10.7
6 in.							
1000 psia							
500 °F							

2.58

BNWL-1084

FIGURE 2.35. Comparison of Plate Loading Waveforms

Conclusions. The conclusions drawn from the results of analyses and experiments described in this section are:

- 1) The results verified the hypothesis that nitrogen solubility in the water was high enough to cause the long-subcooled blowdown regime observed in previous subcooled tests.
- 2) The prediction of subcooled decompression by the WATER HAMMER code gave reasonable agreement with experiment on general rate of decompression, but failed to predict some higher frequency components, which appear to be related to two or three dimensional effects.
- 3) The forces measured on a perforated plate were less than predicted (by the WATER HAMMER-SAMIS-RESPON method) during the subcooled decompression.
- 4) FLASH2 predictions for the saturated portion of a two volume vessel and with no dissolved nitrogen led to slightly different orifice coefficients than previously reported.
- 5) FLASH2 predicts that the liquid level should pass through the internal perforated plate 6 to 20% later than the measured time.
- 6) The fluid state after subcooled decompression remained at saturated equilibrium with experimental accuracy.

Two-Dimensional Effects During Subcooled Blowdown -

N. P. Wilburn

Analysis of the forces applied to the internals of a reactor vessel during the coolant blowdown phase of a postulated accident is required to assure that no unexpected changes in arrangement will occur in the reactor core. Without this assurance of known geometry, there would be serious uncertainty in the calculations of the effectiveness of emergency core cooling systems. The stresses and other effects of the transient forces on vessel internals can be calculated from

knowledge of the pressure as a function of time for all points in the reactor vessel. When the large size and complexity of arrangement of the internals of current reactor vessels are considered, the adequacy of the assumption of a one-dimensional model is open to question. All existing computer codes for the vessel pressure history employ this simplification.⁽¹³⁻¹⁶⁾ For example, WHAM⁽¹⁶⁾ and its predecessor WATER HAMMER⁽¹⁴⁾ solve the single dimension wave equation by the wave superposition and area change reflection principles. This relatively simple approach works rather well in some cases,⁽¹⁷⁾ showing that the assumptions leading to the simple one-dimensional wave equation are good in these cases. However, in other cases the results are not too satisfactory.⁽²⁾ In asking the question, why would the comparisons of the LOFT data⁽¹⁷⁾ with one-dimensional predictions be so much better than those at CSE, one is lead immediately to the possibility that multidimensional effects are occurring in the larger CSE vessel. Confirmation of this possibility was sought in the CSE data. Since all of the pressure gages in the CSE vessel are located on the periphery, radial differences could not be detected. Hence another approach was needed. If radial components of pressure were present, the frequencies corresponding to radial vibrations should be seen in the pressure data taken at the periphery.

The frequencies to be expected can be found approximately by the following simplified two-dimensional analysis. Assume the vessel can be approximated by a cylinder of length L and radius a_0 which has a leak at $Z = 0$ of radius a_1 (See Figure 2.36). By an approach similar to that used before,⁽¹⁴⁾ one can assume that the pressure H is described as a function of position and time by two-dimensional wave equation in cylindrical coordinates:

$$\frac{\partial^2 H}{\partial r^2} + \frac{1}{r} \frac{\partial H}{\partial r} + \frac{\partial^2 H}{\partial z^2} = \frac{1}{c^2} \frac{\partial^2 H}{\partial t^2} \quad (1)$$

TWO DIMENSIONAL APPROXIMATION TO CSE SIMULATOR VESSEL

FIGURE 2.36. Two Dimensional Approximation to CSE Simulator Vessel

where r = radius, Z = axial distance, t = time, and C = sound velocity.

If the velocity of the fluid is taken as zero at the walls everywhere except at the leak and given there as a function of time, and if the initial conditions of the pressure distribution are given, the problem is completely specified and can be solved in principle.

However, the boundary condition at the leak poses difficulty in obtaining a solution by separation of variables. If, however, a_1 is assumed vanishingly small (in the CSE vessel $a_0/a_1 \sim 40$), then an approximate solution can be obtained. The separated equations are:

$$\text{assuming } H = R(r) Z(z) T(t), \quad (2)$$

where R, Z, T are unknown arbitrary functions,

$$\frac{d^2 R}{dr^2} + \frac{1}{r} \frac{dR}{dr} + \omega^2 R = 0 \quad (3)$$

$$\frac{d^2 Z}{dZ^2} + \gamma^2 Z = 0 \quad (4)$$

$$\frac{d^2 T}{dt^2} + k^2 C^2 T = 0 \quad (5)$$

where $\gamma^2 = k^2 - \omega^2$ (γ, k, ω are the eigenvalues of the system).

We expect solution of the form (J_n = the n th order Bessel function)

$$H(r, Z, t) = J_0(\omega_j r) e^{ik_j n t} e^{i\gamma_n Z} \quad (6)$$

The γ_n are determined from the boundary conditions at the ends of the cylinder

$$\frac{\partial H}{\partial Z} = 0 \Rightarrow \gamma_n = \frac{n\pi}{L} \quad (7)$$

and the ω_j from the conditions at a_o

$$\frac{\partial H}{\partial r} = 0 \Rightarrow \omega_j a_o = \{\text{roots of } J_1(\omega_j a_o) = 0\} \quad (8)$$

$$= 0, 3.831, 5, 135 \dots$$

Thus, the fundamental radial frequency contribution would be

$$f = \frac{3.831 C}{2\pi a_o} = 1290 \text{ Hz}$$

where the value of 1.725 ft is taken for a_o (radius of CSE vessel) and 3651 ft/sec is taken for C (velocity of sound in water at 500 °F). This frequency can be compared with some CSE experimental values shown below:

<u>Run</u>	<u>Frequency Noted</u>
B-19B	850 Hz
B-20B	1050 Hz
B-21	1000 Hz

The above experimental frequencies all are smaller than predicted by the simple analysis. However, when the solution to the more complicated situation is found, the arguments of the Bessel function may be different, and this will affect the predicted frequency.

It is planned during the next quarter to attempt a solution to the more complicated problem with the velocity specified for $0 \leq r < a_1$, at $z = 0$. It appears after preliminary consideration that a solution can be obtained using transform methods. A probe is being designed to be installed in nozzle "O" opposite nozzle "B" in the CSE simulator from which blow-downs are to be made in the next series of runs. The probe will contain two pressure gages, one next to the wall and one near the center. According to the simplified analysis given

above, the ratio of the amplitudes of the first two radial modes at the center and at the wall would be

$$\frac{1}{J_0(3.831)} = -2.5, \text{ and } \frac{1}{J_0(5.135)} = -7.55,$$

which means such a probe should have no trouble picking up a pressure difference between these two points. Also, two pressure gages in the vessel wall will be logged at twice the normal high speed multiplexer rate (i.e., at 6000 points per second) in order to give assurance that all the higher frequencies present are recovered.

Reactor Simulator Vessel Defect Indication

Routine Surveillance. Acoustic monitoring techniques are being used to detect acoustic emission signals from the pressure vessel. Such signals may be associated with propagation of the defect region in the bottom head. Monitoring is begun after the coolant blowdown and is continued for roughly one hour. The thermal stresses associated with heat transfer from the steel vessel into the residual water reach a maximum during this interval. No acoustic signals have been detected which can be attributed to defect propagation.

After each blowdown, the defect indication region is given an ultrasonic test to detect any enlargement or change in characteristics. No changes were found by these tests during the current set of experiments.

Defect Propagation Prediction - V. B. Watwood

A method has been devised to use existing finite element codes to predict crack growth rate or the possibility of unstable crack growth of a defect in an axisymmetric body or a two-dimensional planar body. The predictions are based on existing concepts in elastic fracture mechanics.⁽¹⁸⁾ The

primary difficulty in applying these concepts is one of determining the correct value of the stress intensity factor, K , which occurs in the formulation of elastic fracture mechanics.

This work was to provide a means of evaluating the behavior of the laminar type defects⁽¹⁹⁾ which are known to exist in the CSE reactor simulator vessel head, near nozzle "C".

The method of evaluating K is described in detail in a report,⁽²⁰⁾ so only a brief description will be given here. A direct method of computation of K was first tried using the prediction of the stress components or displacements as supplied by one of the existing finite element codes. It was found that accuracy was poor unless extremely small elements are used near the crack tip. Such small elements would have required extensive program modifications before the method could be applied to the CSE vessel configuration. Therefore, the direct method was discarded in favor of a procedure which computes directly the strain energy release rate, G , which is equivalent to K . The method consists of computing the strain energy for two slightly different crack lengths and employing numerical differentiation to determine G . The accuracy is quite good for a relatively coarse finite element mesh for all sample problems.

The method has been used in conjunction with the pressure loads imposed on the CSE vessel with a low value of K indicated. The thermal loading condition is now being checked.

PARTICLE TRANSPORT TO SURFACES BY CONDENSING STEAM -

L. C. Schwendiman, J. M. Hales

The previously derived equation describing aerosol transport to a vertical wall in a laminar, condensing-steam boundary layer⁽²¹⁾ was solved using hybrid and digital computer techniques. The solutions are summarized in Figure 2.37, which shows aerosol concentration profiles for various conditions of temperature and particle size.

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FIGURE 2.37. Concentration Profiles for Aerosol Moving Through a Condensing-Steam Boundary Layer

Using information obtained from the solutions to the total problem, one can compute energy, steam, and particle transport to the wall, and apply these results to the prediction of pressure suppression and aerosol removal by natural-process effects. Equations relating the computed results to wall fluxes are:

Steam flux at wall:

$$\dot{m}_w = \frac{C \mu_\infty \xi' (\Delta W)}{x^{1/4} Sc} \left(\frac{1}{W_i - 1} \right), \quad (1)$$

Aerosol flux at wall:

$$\dot{m}_a = \frac{C}{x^{1/4} \rho} \left[- \frac{N \mu_\infty \Delta W \xi'}{Sc} \left(\frac{1}{1 - W} + \frac{1}{W + 3.729} \right) - \frac{\beta \rho N'}{\phi_\mu^2} \right], \text{ and} \quad (2)$$

Energy flux at wall:

$$\dot{m}_h = - \frac{C}{x^{1/4}} \left[\frac{\lambda \mu_\infty (\Delta W)}{Sc(1 - W_i)} \xi' + \frac{C_p \mu_\infty (\Delta T) \tau'}{Pr} \right]. \quad (3)$$

Equation (1) may be used to calculate the average fluid velocity moving toward the wall. A simple estimation of particle deposition can be obtained by assuming aerosol motion to follow that of the bulk fluid. Particle deposition rates estimated by this one-dimensional approach were compared to those computed from the boundary layer model using Equation (2). A comparison of these results is given in Figure 2.38.

A theoretical investigation of aerosol deposition through a turbulent boundary layer was started. From this it was concluded that a one-dimensional diffusional model of the form

$$\bar{v} \frac{dT}{dy} = [\epsilon + D] \frac{d^2 T}{dy^2} \quad (4)$$

would describe deposition adequately.

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FIGURE 2.38. Ratio of Aerosol Deposition Rates Calculated by Boundary-Layer Model to Deposition Rates Calculated by One-Dimensional Model

Equation (4) was solved for conditions matching those of the Containment Systems Experiment Run A-1, by estimating the eddy diffusivity (ϵ) and assuming a smooth deposition surface. Calculated deposition rates were smaller than those observed experimentally, indicating the estimated values of ϵ were too low. This result should be expected in view of roughness of the containment surface caused by water ripples and droplets.

Nomenclature

$$C = (g/4 v_{\infty}^2)^{1/4}$$

C_p = specific heat

D = particle diffusion coefficient

D_{AB} = mutual diffusion coefficient of air and water

F = transformation variable defined in terms of $F' = \frac{u\phi}{\pi 4 C^2 v_{\infty}}$

g = acceleration of gravity

i = subscript denoting liquid-vapor interface

k = thermal conductivity

$\dot{m}_a$ = flux of aerosol at interface

$\dot{m}_h$ = flux of energy at interface

$\dot{m}_w$ = flux of water at interface

N = particle population density

Pr = Prandtl number, $\frac{C_p \mu}{k}$

Sc = Schmidt number, $\frac{\nu}{D_{AB}}$

T = temperature

u = mass-average velocity in x-direction

v = mass-average velocity in y-direction

W = mass fraction of water

x = direction down plate

y = direction normal to plate

$\beta = D\phi$

δ = liquid film thickness

η = similarity transform variable

λ = latent heat of vaporization

μ = dynamic viscosity

ν = kinematic viscosity

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

T = particle concentration

ρ = density

$$\tau = \frac{T - T_i}{T_\infty - T_i}$$

$$\phi = \mu/\mu_\infty$$

$$\phi_c = C_p/C_{p\infty}$$

$$\phi_{cgv} = \frac{C_{pw} - C_{pa}}{C_{p\infty}}$$

$$\phi_k = k/k_\infty$$

∞ = subscript denoting bulk conditions

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3. PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

CRACK DETECTION IN PRESSURE PIPING BY ACOUSTIC EMISSION - D. O. Hunter, P. H. Hutton, and H. N. Pedersen

Introduction

New methods for detecting formation and/or growth of flaws in nuclear reactor pressure boundaries is of continuing interest to the nuclear industry to further enhance safety. One such method under development by Battelle-Northwest at the Pacific Northwest Laboratory involves detection of "acoustic emission" or elastic waves produced in solids by energy released as the solids deform and fracture. The objective of the program is to develop a system to provide continuous monitoring. The work is sponsored by the USAEC's Division of Reactor Development and Technology and has been in progress since February, 1966.

This development program is related to DRDT sponsored work at Phillips Petroleum Co., Idaho Test Site, concerned with plant application aspects of developed technology. It also has potential application to the LMFBR program.

Task 1 - Acoustic Emission Signal Analysis and Characterization

Previously reported study of acoustic emission wave propagation in flat plates indicated that detection of the shear wave mode offered advantages of a sharp, short detected signal. This was demonstrated through the use of shear sensors mounted on the edge of test specimens. This technique (edge mounting), however, is obviously not directly applicable to emission measurements on a real structure. Work is in progress to devise a practical means of adapting shear wave detection to field applications.

The detection efficiency of a ~ 700 kHz shear PZT-4 receiver crystal has been investigated when transmit energy in a 1/2 in. steel plate at 20 in. was (1) horizontally polarized shear, (2) vertically polarized shear, or (3) compressional, all relative to the plane of the plate. Parameters of the experiment pertained to the coupling medium and the angle at which the receive crystal is aligned relative to the plate.

Figure 3.1 shows an illustration of the crystals' alignment relative to the plate, and directions of propagation and crystals' polarization.

Figure 3.2 shows that a brass coupling wedge will improve reception of a horizontally polarized shear wave by a factor of about 2 compared to coupling the same receive crystal directly to the plate.

Figure 3.3 shows that a brass coupling wedge will improve reception of a vertically polarized shear wave by a factor of at least 2 compared to coupling the same receive crystal directly to the plate. Figure 3.3 also indicates improved vertically polarized shear wave propagation efficiency of about 2 compared to horizontally polarized shear waves as seen in Figure 3.2.

Figure 3.4 shows that the angle of refraction, as coupled through a brass wedge will affect the amplitude of received signal. A slightly higher amplitude signal is extracted at 34.5° than at 45° . Both, however, are higher amplitude than the received signal when coupled directly to the plates' surface.

Figure 3.5 shows the detection of shear waves as a result of compressional waves transmitted from the plates' end. Higher amplitude signals result from direct coupling to the surface than from coupling at about 31.5° (close to the angle of "trailing shear waves" in steel as a result of compressional waves grazing the surface).

FIGURE 3.1. Test Plate, Crystals, and Directions of Propagation and Wave Motions

Time Base: 50 microsec/cm
Gain: 2A

FIGURE 3.2. Detection of Horizontally Polarized Shear Waves Using a Horizontally Polarized Shear Crystal
Top: Using a Brass Coupling Wedge at $\sim 34.5^\circ$
Bottom: Coupled Directly to the Plate's Surface

Time Base: 50 microsec/cm
Gain: A

FIGURE 3.3. Detection of Vertically Polarized Shear Waves
Using a Vertically Polarized Shear Crystal
Top: Using a Brass Coupling Wedge at $\sim 34.5^\circ$
Bottom: Coupled Directly to the Plate's Surface

Time Base: 50 microsec/cm
Gain: A

FIGURE 3.4. Detection of Vertically Polarized Shear Waves
Using a Vertically Polarized Shear Crystal
Top: Using a Brass Coupling Wedge at $\sim 45^\circ$
Middle: Coupled Directly to the Plate's Surface
Bottom: Using a Brass Coupling Wedge at $\sim 34.5^\circ$

Time Base: 50 microsec/cm
Gain: 2A

FIGURE 3.5. Detection of Compressional Waves, Mode-Converted to Vertically Polarized Shear Waves, Using a Vertically Polarized Shear Crystal
Top: Using a Steel Coupling Wedge at $\sim 31.5^\circ$
Bottom: Coupled Directly to the Plate's Surface

Figure 3.6 shows the same information as in Figure 3.5 except that oscilloscope time base has been reduced from 50 microsec/cm to 10 microsec/cm. Notice the existence of repetitive events. These are the "trailing shear waves" and are separated from each other by

$$\Delta t = \frac{d}{V_S \cos \theta} - \frac{d}{V_L} \tan \theta \approx 3.3 \text{ } \mu\text{sec}$$

where

d = plate thickness = 0.5 in.

V_S = shear velocity $\approx 127,000$ in./sec

V_L = compressional velocity $\approx 236,000$ in./sec

$$\theta = \sin^{-1} \frac{V_S}{V_L}$$

The first pulse in each case is the compressional wave which has traveled the entire 20 in. distance at compressional

Time Base: 10 microsec/cm

Gain: 2A

FIGURE 3.6. Detection of Compressional Waves, Mode-Converted to Vertically Polarized Shear Waves, Using a Vertically Polarized Shear Crystal
Top: Using a Steel Coupling Wedge at $\sim 31.5^\circ$
Bottom: Coupled Directly to the Plate's Surface

wave velocity. Subsequently received signals are a combination of waves. That is, part of the distance has been traveled at compressional wave velocity and part by shear wave velocity.

Figure 3.7 shows that no improvement is to be expected over direct coupling by using a brass wedge if the objective is detection of "shear trailing waves" resulting from surface grazing compressional waves.

Conclusions drawn from this experiment are:

- A brass coupling block will improve reception of initial components (rays traveling nearly parallel to the surface) of shear wave energy, either horizontally or vertically polarized, thus enabling more accurate source location through triangulation.
- Some signal optimization may be obtained by selection of an optimum wedge angle.
- Vertically polarized shear waves are received with greater

Time Base: 10 microsec/cm
Gain: 2A

FIGURE 3.7. Detection of Compressional Waves, Mode-Converted to Vertically Polarized Shear Waves, Using a Vertically Polarized Shear Crystal
Top: Coupled Directly to the Plate's Surface
Bottom: Using a Brass Coupling Wedge at $\sim 34.5^\circ$

efficiency than are horizontally polarized shear waves.

- Energy of compressional wave origin is received as shear waves with greater sensitivity by direct coupling than by using either the steel or brass coupling block. Although the receive crystal detects wave motion which is primarily vertically polarized shear, the energy has traveled most of the way at compressional velocity. Better coupling efficiency through the interface of a plate and a steel wedge should improve signal detection relative to that directly coupled to the plates' surface.

Since one of the primary objectives in an acoustic emission detection system is to enable location of the emission's source, coupling parameters must be optimized for the initial received rays which are those traveling parallel to the surface.

The test results described above show that the low propagation velocity of brass better enables reception of emission ray components traveling more nearly parallel to the plane of the plate. In addition, the brass also more nearly approximates the acoustic impedance of the steel and the coupling transducer than do other low velocity materials such as water or plexiglas, thus improving transfer from plate to transducer.

Other materials may improve on brass, but some of these (e.g., gold and platinum) would be too expensive. Two relatively inexpensive materials, which have low propagation velocities and fairly high acoustic impedance, are Zircaloy and silver. To determine the merit of coupling wedges for acoustic emission detection, wedges made from brass, Zircaloy, and silver are being fabricated for evaluation at their respective critical angles for shear waves and surface waves propagating in a 1/2 in. steel plate.

The wedge experiment (using brass only) will be extended to include two steel pipes of different diameters and wall thicknesses. For this purpose, brass adapter plates have been fabricated for coupling the flat wedge to the curved pipe surface.

Recent tests involving 1-1/2 in. thick carbon steel showed evidence of a distinction between signals generated by deformation and those by fracture. Those signals which appear to be related to deformation are lower amplitude and much shorter in duration than the signals from fracture. This correlates with an earlier similar observation from a pipe burst test performed at the Battelle-Columbus test site. Figure 3.8 shows a sample of the observed results. It is suspected that the appearance of this distinction may be a function of using high frequency sensors (about 1 MHz) which are responding differently to the variation in energy pulse period that could be expected

Typical Ductile Fracture Signal

3 1/2 Volts @ ≈ 120 kHz

0.0005 sec/cm Sweep Rate

0.2 Volts @ 25 kHz

0.0002 sec/cm Sweep Rate

Typical Deformation Signals

0.4 Volts @ 25 kHz

0.0002 sec/cm Sweep Rate

FIGURE 3.8. Acoustic Emission from Deformation and Fracture - Steel Pipe Burst Tests

from deformation versus fracture. Controlled tests are planned to confirm this observation.

Work is in progress to investigate acoustic emission response from deformation and fracture of 304 stainless steel as a function of temperature. The fundamental question is whether this material still produces detectable emission at elevated temperature. Similar tests will be conducted at room temperature, 600 °F, 900 °F, and 1200 °F. The work is being sponsored by the Fast Flux Test Facility Program.

Task 2 - High Temperature Transducer Development

Design of the thickness poled, surface mounted, PZT-5A sensor used routinely for test work has been modified to produce a sensor with its major resonance at about 1 MHz. The purpose of this modification is to enhance detection of acoustic emission in the low MHz range by minimizing interference from lower frequency sensor response. Some loss in sensitivity relative to the lower frequency sensor was incurred but the sensitivity is still adequate for test work.

Task 3 - System Analysis Study

No further work.

Task 4 - Monitor System Development

No further work.

Task 5 - Cooperative Work with Phillips Petroleum Company

Verbal liaison only.

Task 6 - Application Engineering

Application of acoustic emission detection locally to investigate potential slow crack growth in process piping by fatigue has continued. The particular sections of piping are known to experience high local stresses during operation. Four of eight locations have been monitored through a pressurization

cycle to about 115% of current normal operating pressure. In three cases, indications of very limited crack activity were detected. The size of the cracks are below the resolution limit of ultrasonic inspection used for follow-up examination of the emission source area.

The Containment Systems Experiment Program has continued to use acoustic emission monitoring for surveillance of known flaws in their simulator vessel heads. Four tests have been monitored during this quarter. Data from the first two have been analyzed with no significant indications. Data from the last two are still in the process of analysis.

General

A paper titled "Acoustic Emission - A New Tool for Evaluating Structural Soundness" was presented at the Seventh Symposium on Nondestructive Evaluation of Components and Materials in Aerospace, Weapons Systems, and Nuclear Applications at San Antonio, Texas, April 23-25, 1969.

Work Planned for Next Quarter

1. Continue work on adaptation of shear sensor to continuous surfaces.
2. Investigate maximum detection distance at 1 to 2 MHz and the effect of temperature on attenuation of the propagated signal.
3. Design a sharper (30 to 36 dB/octave) filter for use in test work.
4. Initiate study of sensor couplants for extended service at 600 to 700 °F.
5. Endeavor to authenticate distinction between deformation and fracture signals in carbon steel.

4. ENVIRONMENTAL STUDIES

SIMULATION MODELING OF EXPECTED THERMAL GENERATION

IN SELECTED RIVER SYSTEMS - R. T. Jaske and D. E. Peterson

Operations research with the COL HEAT thermal simulation model to investigate the effects of future nuclear power plants on temperatures of Ohio River Basin streams was initiated during the month of April.

During the report period the following accomplishments were made:

- The collection of basic data associated with the Ohio River Basin study was continued. Compilation of these data into card format is largely complete. First and second test simulations of 1964 daily average temperatures on the Allegheny, Monongahela, Muskingum, Kanawha, Big Sandy, Kentucky, Wabash, and Ohio Rivers are scheduled for May.
- In order to check the operation of the COL HEAT simulation model with real data, simulation of daily average temperatures on the Illinois River from its origin at the confluence of the Des Plaines and Kankakee Rivers to the Mississippi River was carried out for the period July through December 1968. The computed and observed temperatures at Starved Rock Dam are illustrated in Figure 4.1. The observed temperatures in this study were obtained from four thermographs placed specifically for studies of the thermal regimen of the Illinois River at Dresden Island Dam, Starved Rock Dam, La Grange Dam, and at Alton Dam (Alton Dam is on the Mississippi River near its confluence with the Illinois River).

FIGURE 4.1. Comparison of Observed and Computed River Temperatures for the Illinois Waterway for the Period July-December, 1968. (Data from AEC Instruments)

- A study of the hourly temperature conditions on the Phelps County Canal (Nebraska) were continued. The purpose of this study is to test whether regional weather and solar radiation data can be used in the simulation of hourly average canal temperatures which could be useful for heat dissipations. The nearest data stations are in this case approximately 100 miles from the center of the area of study. The results to date have not attained the desired level of accuracy. Consequently, the study is continuing with the object of developing approaches which are completely satisfactory.

5. FIXATION OF RADIOACTIVE RESIDUES

SPRAY SOLIDIFICATION

LABORATORY STUDIES

Chemical Additives for Processing PW-4m - J. E. Mendel

Results of laboratory tests with PW-4m revealed that melting points were generally lowered as much as 50 °C as the M/P ratio was decreased from 2.75 to 2.25. Since the tendency for phase separation in the product was greater at an M/P ratio of 2.25, a compromise ratio of 2.50 was adopted for further studies. The effect of composition upon melting point at an M/P ratio of 2.50 is shown in a ternary plot (Figure 5.1). The coordinates are fission products, iron, and sodium, all expressed as cation monoequivalents. The compositions shown were obtained by varying the amounts of iron and sodium added to PW-4m (PW-4m as used in these studies contained 1.68 FP M⁺, 0.15 Fe M⁺, and 0.34 Na M⁺). Although sodium was used as the alkali metal additives here, other studies with both PW-4m and PW-1 have shown that an equimolar mixture of sodium and lithium yields slightly lower melting points, hence the mixture was used in WSEP Run SS-9. Increases in fission product content cause both melting point and viscosity to increase.

The fission product content in Run SS-9 product was 25 wt% as oxide. The chemical additives used in Run SS-9 were nominally 1.10M Fe, 0.75M Li and 0.75M Na (where M equals moles per liter at a waste concentration of 370 liters/tonne) at an M/P ratio of 2.50. In the actual run the iron additive concentration was only 0.9M. The waste contained 0.2M Al from the Purex 1WW solutions used for feed makeup and this was treated as an iron substitute. In Run SS-9 the melting point and viscosity were high enough to cause slow

FIGURE 5.1. PW-4m Calcine Melting Points at an M/P Ratio of 2.50

draining of the melter. As can be seen in Figure 5.1, increasing the sodium content would yield somewhat lower melting points and permit incorporation of even larger quantities of fission product. Use of more sodium was considered inadvisable because 1) leachability of the product increases and 2) the melt develops a tendency to separate into two phases.

Corrosion Studies - R. F. Maness

Specimens of 304L and 310 stainless steel, and mild steel were exposed in 24 to 50 hr tests to PW-4m spray calciner melt held at temperatures ranging from 700 to 1000 °C. The corrosion rate of 310 at 950 and 1000 °C was about 145 and 170 mils/year, respectively. A 1/4 in.-thick 304L specimen dissolved in melt held at 1000 °C in less than 24 hr, whereas

a specimen exposed at 950 °C corroded at a rate of 190 mils/year. The corrosion rate of mild steel in melt held at 700 °C was 340 mils/year.

ENGINEERING STUDIES

Development Spray Solidification Runs - J. D. Moore

Six spray solidifier tests were carried out to develop a PW-4m flowsheet which could be used in the WSEP radioactive demonstrations. It was desired to develop a low viscosity melt and a homogeneous solid with a melting point below 1000 °C. Retention of ruthenium in the product was also desired. A summary of run operating conditions and results is given in Table 5.1.

Solid NaPO_3 flux was added to calcined low acid (see Table 5.2 for feed compositions) PW-4m in the melter during Run DSC-25. Ruthenium release to the off-gas was limited to only 2%. Laboratory tests with the melt product, however, showed that a rare earth rich phase with a melting point greater than 1200 °C precipitates from the molten state after about 4 hr. This precipitate could plug both melter discharge valves if the melt were allowed to set in the melter for several hours without mixing.

Homogeneous melts were formed in DSC-26 and -27 when iron was added to the feed. Ruthenium volatilization increased to 31% in DSC-27, however, as it was necessary to add phosphoric acid flux to the feed, thus increasing the feed acidity.

Aluminum (0.2M) was substituted for 0.2M iron in Runs DSC-28 through -30. The substitution was made since the aluminum is contained in the Purex 1WW used for feed

TABLE 5.1. Summary of Development Spray Solidification Runs

	DSC-25	DSC-26	DSC-27	DSC-28	DSC-29	DSC-30
Feed type	PW-4m	PW-4m	PW-4m	PW-4m	PW-4m	PW-4m
Feed concentration, liters/tonne	378	458	493	522	548	573
Additives	NaPO ₃ , solid	NaPO ₃ , solid H ₃ PO ₄ , Fe	Fe, NaPO ₃ , H ₃ PO ₄	Fe, Al, NaPO ₃ , H ₃ PO ₄	Fe, Al, NaPO ₃ , Li, H ₃ PO ₄	Fe, Al, Na, Li, H ₃ PO ₄
Feed free acid, H ⁺ _M	0.037	2.92	4.09	4.09	5.59	5.94
M ⁺ /P	2.2	2.25	2.25	2.25	2.25	2.5
Feed consumed, liters	52.6	150.2	298	125	84.5	100
Average feed rate, liters/hr	12.1	15.8	14.9	16.3	10.6	14.3
Calciner furnace temperature, °C	700	700	700	700	760	700
Calciner internal temperature near spray, °C	550	550	495	490	600	535
Calciner, vacuum, inches of water	8	8	8	8	8	8
Atomizing gas	air	air	air	nitrogen	air	air
Atomizing gas flow, scfm	6.7	6.5	6.8	7	6.5	6.6
Filter blowback pressure, psig	40	40	36-44	40	40	30-65
Filter pressure drop, end of run, inches of water	4.3	6	4.5	3.3	2.5	1.3
Melter furnace temperature, °C	1150-1175	1100-1225	1150-1175	1175-1200	1150-1200	1150-1220
Product receiver furnace temperature, °C	unheated	900	unheated	unheated	unheated- 800	unheated
Product collected, kg	19	46	93.6	35.4	24.9	23
Product density, g/cm ³	3.17	3.35	3.3	3.27	3.32	3.51
Product concentration, liters/tonne	43	42	47	45	49	38
Particulate DF (feed to condensate)	860	2250	5400	>800	2700	1200
Ruthenium volatilized, % of total ruthenium	2	7	31	60	29	19

TABLE 5.2. PW-4m Feed Composition for Development Spray Solidifier Runs Basis: 387 liters/tonne concentration

<u>Chemicals</u>	
Fe ⁺³	0.05M
Cr ⁺³	0.012
Ni ⁺²	0.008
Al ⁺³	0.001
ZrO ⁺²	0.143
Na ⁺	0.342 ^(a)
SiO ₃ ⁻²	0.010
PO ₄ ⁻³	0.003
<u>Fission Products</u>	
MoO ₄ ⁻²	0.130
Sr ⁺²	0.036
Ba ⁺²	0.041
Rb + Cs (used K ⁺)	0.092
(Y + RE) ⁺³	0.274
Ru ⁺⁴	0.0164 ^(b)
Ru ⁺⁴ (used Fe ⁺³)	0.066
Rh (used Co ⁺²)	0.013
Ag ⁺	0.0016
Cd ⁺²	0.0025
Te (used SO ₄ ⁻²)	0.014
Pd (used Ni ⁺²)	0.043
Tc (used MoO ₄ ⁻²)	0.031
NO ₃ ⁻	1.941
H ⁺	0.037

<u>Additives</u>	<u>DSC-25</u>	<u>DSC-26</u>	<u>DSC-27</u>	<u>DSC-28</u>	<u>DSC-29</u>	<u>DSC-30</u>
Fe ⁺³	--	0.7	0.9	0.7	0.7	0.9
Al ⁺³	--	--	--	0.2	0.2	0.2
Li ⁺	--	--	--	--	0.75	0.75
Na ⁺	2.0	solid add to melter	1.5	1.5	0.75	0.75
PO ₃ ⁻	2.0		1.5	1.5	0.75	--
PO ₄ ⁻³	--	0.96	1.35	1.35	2.1	2.8
OH ⁻	--	--	--	--	0.75	1.5
NO ₃ ⁻	--	2.1	2.7	2.7	2.7	3.3
H ⁺	--	2.88	4.05	4.05	6.3	8.4

a. Nominal PW-4m concentration is 0.1M. Excess is due to using Na₂MoO₄ for molybdenum addition.

b. 0.0123M for DSC-29 and 0.0082M for DSC-30. Nominal PW-4m Ru concentration is 0.082M.

makeup in the WSEP radioactive demonstrations. The melts produced were more viscous than those containing no aluminum, but they dripped freely from the melter weir at temperatures of 1050 to 1100 °C. In DSC-28, the melt foamed excessively, but very little foaming was experienced in the following runs. The solidified melts were dark gray crystalline materials which separated into a glassy and heavier crystalline phase when held without agitation at 1050 °C for 4 to 7 hr. Both phases melted at 1000 °C, or below, however. The phase separation was not evident in the product in the melt receiver pot. The mixing action of the melt in the melter should keep the phases from separating during normal operation.

Attempts to reduce the ruthenium volatilization to low levels (less than 10%) in DSC-28 and -29 were inconclusive. In DSC-28, nitrogen atomizing gas was used in place of air to produce a less oxidizing atmosphere to suppress the formation of volatile RuO_4 . Air leakage into the calciner diluted the nitrogen by a factor of two, however, and 60% of the ruthenium was released to the off-gas. An attempt to reduce ruthenium volatilization by increasing the internal calciner temperatures in the spray area was made during DSC-29. Temperatures were increased approximately 50 to 100 °C to 600 °C by raising the furnace temperature from 700 to 760 °C and reducing the feed rate to 10 liters/hr from the normal 15 liters/hr. Although only 29% of the ruthenium volatilized, this decrease could not be entirely attributed to the higher temperature since only 19% of the ruthenium evolved during DSC-30 when the calciner internal temperature was 65 °C below that of DSC-29. Other potential ruthenium retention methods such as feed acid-stripping and chemical neutralization are being tested.

Batch-filling an unheated storage pot was successfully demonstrated during DSC-27. Melt heated to 1050 °C was drained

from the melter freeze valve into the pot in 5-liter batches. The 8-in. diameter, 304L stainless steel pot was filled evenly and was not corroded by the melt.

Feed flow stability was relatively good with all of the feeds tested. Calciner internal wall deposition was minimal and the off-gas filters were kept clean with a blowback gas pressure of 40 psig. Particulate de-entrainment factors were good, ranging from 800 to 5400 for the six runs.

RADIOACTIVE DEMONSTRATION - W. R. Bond, J. N. Hartley, M. R. Schwab, and W. K. Winegardner

Spray Solidification Run SS-8

During Run SS-8 simulated waste equivalent to 1.8 tonnes of power reactor fuel (20,000 MWd/tonne at 15 MW/tonne) with an equivalent aging time of 1.2 years was processed to produce 195 kg of ceramic product. The 59 liters of radioactive ceramic produced a heat-rate density of 109 W/liter (6500 W total based on analytical data and 6800 W total based on pot calorimetry) in an 8-in. diameter 304L stainless steel melt receiver. The steady-state pot centerline temperature in the air-cooled furnace was 605 °C and the centerline-to-wall temperature difference was 210 °C. These values resulted in an effective thermal conductivity of $1.4 \text{ W/(m}^2)(^\circ\text{C/m)}$ [$0.79 \text{ Btu/(hr)(ft}^2)(^\circ\text{F/ft)}$] for the ceramic product.

The solidifier was fed 850 liters of low acid, sulfate-free PW-1 feed at 473 liters/tonne in 55 hr at an average feed rate of 15.5 liters/hr. The feed containing 1,800,000 Ci of radioactivity including 14,000 Ci of radioactive ruthenium was reduced in volume by a factor of 14.4 or 11.5 on a 378 liters/tonne basis. A summary of run data from SS-8 is presented in Table 5.3.

TABLE 5.3. Summary Data for WSEP Spray Solidifier Runs

<u>Operational Data</u>	<u>SS-8</u>	<u>SS-9</u>
WSEP Run	20	21
Date	2/69	3/69
Operating Mode	A	A
Feed Type ^(a)	PW-1	PW-4m
Feed, liters/tonne	473	630
Pot Diameter, in.	8	8
Pot Material	304-L	310
Total Run Feed Time, hr	55	52.2
Average Feed Rate, liters/hr	15.5	15.2
Total Feed, liters	850	792
Total Feed, tonne equivalent	1.8	1.26
Feed: Solid Volume Ratio	14.4	11.7
Feed: Solid Volume Ratio at 378 liters/tonne Basis	11.5	7.0
Filter ΔP, in. of Water	4-5	5-6

Problems

SS-8 Calciner inlet to melter plugged at end of run; inlet nozzle was cleared mechanically prior to final melter dump.

SS-9 Pneumatic vibrator had to be replaced during the run and resulted in 7.0 hr of feed time interruption.

<u>Filled Pot Data</u>		
Total Curies in Feed to Pot ^(b)	1,800,000	2,190,000
Total Curies of Ru in Feed to Pot ^(b)	14,000	10,200
Solid in Pot, liters	59	68
Solid in Pot, kg ^(c)	195	195
Solid in Pot, liters/tonne	33	54
Heat in Pot, W ^(d)	6800	7200
Equivalent Aging Time, years	1.2	2.2
ε Temp in Air-Cooled Furnace, °C	605	594
ε to Wall Δ Temp., °C	210	242
Effective Thermal Conductivity, W/(m ²) (°C/m) [Btu/(hr) (ft ²) (°F/ft)]	1.4 [0.79]	1.14 [0.66]
Welded Lid	Yes	Yes
Radiation Reading, R/hr at 6 in.	NA	26,000

NA Not Available.

a. See Table 5.2 for feed composition.

b. By feed analysis.

c. Calculated from lab density and rodded volume.

d. By pot calorimetry data.

At the end of SS-8, a calcine plug formed above the melter inlet and it was necessary to clear it mechanically prior to the final melter dump. Indications of plugging were noted half-way through the run when the calcine fall into the melter became erratic and progressively worsened until no calcine at all was observed falling at the end of the run. The plug was broken by rodding and the bulk of the calcine was brought down using the calciner vibrator. Approximately 75% of the calcine for the final melter dump had been retained in the calciner. The calcine appeared free-flowing and no chunks were observed. After the run, the melter was disconnected from the spray calciner and both were inspected to determine the cause of the plugging. Inspection of the inlet to the melter revealed it to be clear. There was, however, some calcine deposition (1 to 2 kg) in the conical bottom section of the calciner directly below the barrel, but it did not appear to represent enough of an obstruction to have caused a plugging problem. The feed flow during the last half of the run was somewhat erratic as indicated by fluctuations in the calciner internal temperatures and may have been responsible for the deposition in the calciner cone. Also, scale in the calciner barrel appeared more pronounced than in the previous inspection (prior to SS-7), but was not present to the extent that special cleaning of the barrel was required.

The filter pressure drop averaged about 4 to 5 in. of water throughout the run indicating that the blowback system performed quite satisfactorily.

Prior to the run, the simulated PW-1 waste including the phosphoric acid additive was acid stripped in the waste evaporator (TK-113). After the stripping operation and the addition of the sodium and lithium hydroxide additives,

the total acidity of the feed had been reduced from a nominal 9M to 1.6M (based on 378 liters/tonne). As shown in Figure 5.2, despite the factor of five reduction in feed acidity, 35% of the ruthenium in the feed to the solidifier was volatilized during SS-8. Although this represents a reduction from the 50 to 70% volatility experienced in previous spray solidifier runs with the PW-1 flowsheet, it does not bear out the strong dependence of ruthenium volatility on feed acidity found in the "cold" development spray solidifier studies.*

During SS-8, 5% of the nonvolatile radioactive species (other than ruthenium) in the feed accumulated in the evaporator (TK-113). Since condensate samples from the solidifier off-gas condenser (E-111) indicated that less than 0.1% of the radioactive species other than ruthenium were being entrained from the solidifier during the run, it was suspected that feed leaked directly into the evaporator during the run through a control valve that separates the evaporator from the solidifier feed system. A review of the operating data indicated that the valve had not been operated until just prior to Run SS-7. Thus, the high entrainment previously* reported for the spray solidifier is probably due in part to leakage through the valve and the flush of the feed line to the evaporator which occurred during that run. Condensate (E-111) sample analyses for Run SS-7 indicate that less than 0.1% of the radioactive species other than ruthenium were entrained from the solidifier during that run.

The cumulative decontamination factor, DF_c , for ruthenium across the WSEP auxiliary evaporator was 1.1×10^1 . Instantaneous decontamination factors, DF_i 's for ruthenium ranged from 5.1×10^2 near the start of the run to 4.4×10^4 after the solidifier feed was off. This, combined with a nearly

FIGURE 5.2. Ruthenium Volatility During Spray Solidification Runs

constant and excessive rate of ruthenium accumulation in the acid fractionator indicated direct gas phase flow of ruthenium from the process condenser to the evaporator condenser as occurred in Run SS-7. The DF_i 's across the evaporator for nonvolatiles represented by $^{144}\text{Ce-Pr}$ averaged greater than 10^6 .

The DF_c across the fractionator was 7.8×10^2 for ruthenium. The DF_i 's for ruthenium averaged greater than 10^3 .

The final condensate stream (fractionator condensate) contained a fraction of original feed activity equal to 5.0×10^{-6} for ruthenium and 1×10^{-9} for nonvolatiles. These values were above 10CFR20 release limits by factors of 1.4×10^4 and 1.8×10^2 , respectively.

Fractional losses of activity by off-gases to the stack were less than 2×10^{-10} for ruthenium and less than 4.6×10^{-13} for gross beta.

The distribution of the feed radioactivity for SS-8 is given in Table 5.4.

Nitrogen analysis of the calciner condensate accounted for only 65% of the total nitrogen volatilized from the solidifier. The remaining 35% remained as oxides of nitrogen which were carried through the WSEP evaporator and fractionator in the gas phase as in previous spray solidifier runs. An additional 10% of the volatilized nitrogen was removed in the WSEP scrubber which contained distilled water, and the remaining 25% of the nitrogen fed to the solidifier escaped

* BNWL-1003, 68-4.

TABLE 5.4. Activity Distribution for Runs SS-8 and SS-9
(Mode A Operation) (a)

	Fraction of Equivalent Feed Radioactivity to Solidifier in Effluent Streams					
	¹⁰⁶ Ru		¹⁴⁴ Ce-Pr		Total Less Ru	
	SS-8	SS-9	SS-8	SS-9	SS-8	SS-9
Through Process Condenser (E-111)	3.9 ⁻¹	5.7 ⁻¹	5.7 ⁻² ^(b)	2.2 ⁻⁴	7.0 ⁻²	2.2 ⁻⁴
Through Evaporator Condenser (E-113)	3.6 ⁻³	6.5 ⁻³	1.2 ⁻⁵	NA	1.2 ⁻⁵	NA
Through Fractionator Condenser (E-115)	4.8 ⁻⁶	2.3 ⁻⁶	1.0 ⁻⁹	NA	1.0 ⁻⁹	NA
Off-Gas to Scrubber	2.5 ⁻⁵	1.8 ⁻⁵	4.3 ⁻⁸	NA	4.3 ⁻⁸	NA
Off-Gas to Stack	<2.8 ⁻¹⁰	<2.8 ⁻¹⁰	NA	NA	<4.9 ⁻¹³	NA
Ratio (Fract. Cond.) Activity to 10CFR20	1.4 ⁵	NA	1.8 ²	NA	1.8 ²	NA
Ratio (stack) Activity to 10CFR20	<6.8 ⁻³	<6.8 ⁻³	NA	NA	<8.2 ⁻⁴	<8.2 ⁻⁴

NA Not Available

a. BNWL-507, 67-2.

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

b. Includes feed radioactivity leak in feed loop direct to evaporator.

from the auxiliary off-gas train to the stack. During the first two-thirds of the run, about 60 liters/hr of fractionator condensate was added as a spray to the top of the calciner condenser to absorb ruthenium and oxides of nitrogen from the solidifier off-gases. However, the condensate addition produced no significant reduction in either the ruthenium concentration or the oxides of nitrogen in the off-gases.

Spray Solidification Run SS-9

During Run SS-9, a simulated waste equivalent to that resulting from the reprocessing of 1.26 tonne of power reactor fuel (45,000 MWd/tonne at 30 MW/tonne) with an equivalent aging time of 2.6 years was processed to produce 194 kg of ceramic product. The 68 liters of radioactive ceramic produced a heat rate density of 106 W/liter (7,300 W total based on analytical data and 7,150 W total based on pot calorimetry) in an 8-in. diameter 310 stainless steel melt receiver. The steady-state pot centerline temperature in the air-cooled furnace was 594 °C and the centerline-to-wall temperature difference was 242 °C. These values indicate an effective thermal conductivity of $1.14 \text{ W/(m}^2)(^\circ\text{C/m)}$ [$0.66 \text{ Btu/(hr)(ft}^2)(^\circ\text{F/ft)}$] for the ceramic product.

The solidifier was fed 792 liters of adjusted PW-4m* feed (additives of 0.75M LiOH, 0.97M NaOH, 1.1M $\text{Fe(NO}_3)_3$ and 2.8M H_3PO_4 based on 378 liters/tonne) at 630 liters/tonne in 52.2 hr. The feed containing 2,190,000 Ci of radioactivity including 10,200 Ci of radioactive ruthenium was reduced in volume by a factor of 11.7, or 7.0 on a 378 liters/tonne basis. A summary of run data for SS-9 is presented in Table 5.3.

* BNWL-889, 68-2.

In general the spray solidifier and its principal components (atomizing nozzle, off-gas filters, and feed control system) performed satisfactorily. Two periods of mechanical difficulty with the pneumatic vibrator were encountered. The feed was interrupted both times with replacement of the vibrator required the final time.

The melt was discharged batchwise to the receiver from the melter via the freeze-drain valve. The PW-4m melt was considerably more viscous than either PW-1 or PW-2 melts and as a result it took 3 to 4 times longer to discharge the melt than in previous runs. Despite the slower discharge rate a uniform fill of the unheated receiver pot was attained.

In addition to demonstrating solidification of PW-4m waste, particular emphasis was placed upon the investigation of the distribution of radioactivity in the decontamination equipment (auxiliaries) located downstream of the spray solidifier. As shown in Figure 5.2, 57% of the ruthenium fed to the solidifier was volatilized and then collected in the evaporator. On the other hand, only 0.022% of the non-volatiles (represented by $^{144}\text{Ce-Pr}$) were entrained from the solidifier. The cumulative decontamination factor, DF_c , across the WSEP auxiliary evaporator ruthenium was 87. Instantaneous DF_i 's for ruthenium ranged from 3.4×10^2 to 1.9×10^3 . Corresponding values for $^{144}\text{Ce-Pr}$ (representing nonvolatiles) ranged from 5.2×10^3 to 1.8×10^4 . Additional data is presented in Table 5.4.

Review of data had suggested that the source of at least part of the ruthenium collected in auxiliary system components during runs was not that fed to the solidifier, but material

plated or trapped on filters, pipe walls, etc. In an attempt to "decontaminate" and qualitatively define the extent of this material, 188 liters of 5.6M nitric acid and 180 liters of 6M nitric acid were fed to the solidifier before and after the radioactive feed portion of SS-9, respectively. Acid flow rate was about 15 liters/hr. A preliminary review of data indicates that ruthenium buildup in auxiliaries during these acid or nonradioactive feed periods was negligible when compared to the radioactive feed portion of SS-9.

Instantaneous ruthenium DF_i 's for the evaporator during the above two acid runs ranged from 2.8×10^4 to 1.9×10^5 . The fact that better decontamination factors are attained without radioactive feed supports the assumption initially based on a nearly linear accumulation of ruthenium in the acid fractionator, that much of the contamination of equipment downstream of the evaporator is the result of a non-condensable gas stream from the solidifier. This gas stream results in the deposition in the fractionator condensate and the aqueous phase of the scrubber of about 1×10^{-5} and 4×10^{-5} of the radioactive ruthenium fed to the solidifier, respectively. During the last 12 hr of the radioactive portion of Run SS-9, fractionator condensate was added as a spray to the process condenser (installed directly downstream of the solidifier and between the solidifier and evaporator). As was the case for SS-8, no obvious scrubbing effect was attained with this technique.

After the final acid run, a 12 hr acid strip of the evaporator bottoms was performed. Data concerning ruthenium volatility during this portion of the run are not yet available.

During SS-9 work was also begun on gas sampling techniques in attempts to provide engineering-scale data that will permit better definition of the type, form, and extent of activity present in the process off-gas stream. Gaseous effluent from the scrubber was sampled for both particulate and volatile forms of activity by routing a side stream of the effluent through first a glass fiber filter and then a 2M NaOH solution. Work is continuing to assure that representative samples are being taken and the following initial results should be regarded as tentative. Analysis of filters revealed that the primary contributor to the "particulate" activity is ^{106}Ru . Preliminary calculations suggest that the order of magnitude for the ^{106}Ru concentration in the process off-gas is $10^{-8} \mu\text{Ci}/\text{cm}^3$, while that for $^{144}\text{Ce-Pr}$ is about a factor of 10 less.

PHOSPHATE GLASS SOLIDIFICATION

LABORATORY STUDIES

Corrosion Studies - R. F. Maness

Crucibles fabricated of chromium and of 23 Cr-77 Al_2O_3 (both deposited by flame spraying) were evaluated in PW-1 phosphate glass melt at 1000 °C. The crucibles (3 in. high, 2 in. OD, 0.160 in. wall thickness) were filled about one-third full with melt and held at temperature for 180 hr. During this interval the crucibles were removed from the furnace on six occasions and cooled to room temperature in still air. No cracking and little corrosion was noted. However, both crucibles were attacked severely when the temperature was increased to 1150 °C for 70 hr. In addition to severe corrosion (about 50 mil wall thickness remaining in the interface

area), both crucibles were cracked. Two hairline cracks were visible in the 23 Cr-77 Al₂O₃ crucible, whereas numerous large cracks were apparent in the chromium crucible.

Specimens of 50 Cr-50 Ni alloy were exposed for 50 to 170 hr to PW-4m phosphate glass melt at 850, 950, and 1000 °C. Corrosion rates were 10, 18, and 69 mils/year, respectively. Similar tests with PW-1 phosphate glass melts gave corrosion rates of 11, 47, and >5000 mils/year, respectively.

Inconel 702 was evaluated as a candidate material (as air-conducting tubing) to control the freeze valve on the phosphate glass melter. A section of tubing was exposed for 162 hr in the vapor phase above a PW-2 phosphate glass melt held at 1000 °C. The specimen was not attacked significantly.

STUDIES ON AUXILIARIES TO SOLIDIFICATION SYSTEMS

Computer Simulation of WSEP - P. C. Owzarski and C. R. Cole

Differential equations describing behavior of nitrogen oxides, nitric acid, and radionuclides in the gas phase have been derived for the three WSEP process condensers, and the fractionator and caustic scrubber. These equations are being solved for various solidifier off-gas compositions.

Results available now show expected decontamination factors for the caustic scrubber acting on Brownian aerosol particles and RuO₄ gas. The relationship derived is

$$DF^{-1} = \exp(-Z/H_G)$$

where:

$$H_G = \alpha \frac{G^\beta}{L^\alpha} S_c^{1/2}$$

and:

Z = tower height

G = gas mass flow rate, lb/hr-ft²

L = liquid mass flow rate, lb/hr-ft²

H_G = height of a gas phase transfer unit

S_c = Schmidt number, kinematic viscosity/particle diffusivity

α, β, γ = constants particular to packing type*

For the WSEP scrubber (1 in. raschig rings, 8 ft tall x 1 ft² cross section) and 38 scfm of air flow, the following equation resulted:

$$DF = \exp(44.2 D^{0.5})$$

where:

D = particle diffusivity, cm²/sec.

This was used to compile the following table:

Caustic Scrubber Decontamination Factors at 25 °C

Particle Diam., microns	D, cm ² /sec**	DF
1.0	2.76 x 10 ⁻⁷	1.02
0.4	8.4 x 10 ⁻⁷	1.04
0.2	2.2 x 10 ⁻⁶	1.07
0.1	6.8 x 10 ⁻⁶	1.12
0.04	3.6 x 10 ⁻⁵	1.30
0.02	1.4 x 10 ⁻⁴	1.69
0.01	5.3 x 10 ⁻⁴	2.77
0.005	2.1 x 10 ⁻³	7.6
0.002	1.3 x 10 ⁻²	154
0.001	5.2 x 10 ⁻²	2.4 x 10 ⁴
RuO ₄ gas	0.073	1.5 x 10 ⁵
(10 ⁻⁴) -H ₂ molecule	(0.7)	

* R. E. Treybol, "Mass Transfer Operations," McGraw-Hill, N.Y., 1955.

** C. N. Davis, ed., "Aerosol Science," Academic Press, N.Y., 1966.

From this table, two conclusions can be drawn: First, the tower will adequately scrub gaseous RuO_4 and second, the tower will not scrub the larger Brownian particles. Therefore, high efficiency filters must be relied upon to remove radionuclide aerosols.

Ruthenium Decontamination During Denitration of SS-8 Feed -
P. C. Owzarski

Excellent decontamination data were obtained prior to Run SS-8 during denitration of the SS-8 feed. Instantaneous ruthenium DF's were calculated for the evaporator based on ^{106}Ru concentrations and are plotted on Figure 5.3. There is an apparent correlation between Ru volatility and overhead acidity. The slope of the curve is about three, which indicates the decontamination factor is proportional to (overhead acidity)⁻³.

INTERMEDIATE AND LOW LEVEL WASTE TREATMENT

MELTER CONDENSATE FROM PHOSPHATE GLASS PROCESS

Development Studies - W. T. McKean

Cold runs with the Distillation Demonstration Unit (DDU) using simulated melter condensate have been completed. Most of the runs were made using simulated feed mixtures described in the previous quarterly report.* In addition,

* BNWL-1003, 68-4.

FIGURE 5.3. WSEP Auxiliary Evaporator Instantaneous Ruthenium Decontamination Factors

one run has been made in which the simulated feed contained ferric and sodium salts at levels corresponding to PG-4 condensate.

H_3PO_4	0.19M
HNO_3	0.455
H_2SO_4	0.5
Ru^{+4}	0.0067
Fe^{+3}	0.35 g/liter
Na^{+}	0.55 g/liter

The evaporator bottoms from this run were considered more viscous than those in runs which did not contain salts. It was necessary to add dilution water to prevent plugging of the dip tubes while removing the bottoms from the evaporator. Ruthenium analysis has not been completed on the nitric and sulfuric acid distillation streams; however, qualitative visual observation of the samples did not indicate abnormal ruthenium evolution relative to the other cold runs discussed in the previous report.*

Corrosion was found on stainless steel fittings on the nitric and sulfuric acid receiving tanks and on the liquid sampler at the base of the DDU condenser. These were replaced and the vacuum system checked to ensure no leaks. No significant corrosion of the stainless steel receiving tanks themselves could be detected by ultrasonic sound measurement of the wall thickness.

* BNWL-1003, 68-4.

EVALUATION OF SOLIDIFIED WASTE PRODUCTS

NONRADIOACTIVE LABORATORY STUDIESProduct Characterization - Continuation of Laboratory
Evaluation of PW-2 Phosphate Glass Samples - J. H. Kleinpeter

The thermal conductivity has been determined for sixteen samples of phosphate glass PW-2 melt that were prepared to study the effect of melt temperature, time at melt temperature, cooling rate, and storage temperature. There appears to be a significant difference between samples stored at 100 °C and those stored at 600 °C. In general, those samples stored at 600 °C tend to be crystalline in structure and tend to have a higher thermal conductivity than those stored at 100 °C.

Table 5.5 shows the average thermal conductivity of the samples as calculated from measurements over a temperature range from 75 to 325 °C after storage at 100 and 600 °C. There is some overlap of conductivities, but in general a dividing line can be drawn between the thermal conductivities for the two storage temperatures. It is possible that interactions of the other variables (melt temperature, cooling rate, and time at melt temperature) are indicated by this overlap. These other variables are listed in the previous quarterly report.*

* BNWL-1003, 68-4.

TABLE 5.5. Average Thermal Conductivity of Nonradioactive Phosphate Glass Samples

<u>Sample No.</u>	<u>Storage Temperature, °C</u>	<u>Average Thermal Conductivity, W/(m²) (°C/m)</u>
2	600	0.96
3	600	0.89
7	600	0.88
9	600	0.82
10	600	0.86
8	600	0.84
15	600	0.68
16	600	0.96
4	100	0.77
5	100	0.73
6	100	0.82
13	100	0.81
11	100	0.58
12	100	0.62
14	100	0.63
17	100	0.57

Dispersibility data determined by W. L. Mularz* has been recalculated and tabulated in a simplified form. These data indicate that there is a strong correlation between particle size distribution and storage temperature.

<u>Sample No.</u>	<u>Storage Temperature</u>	<u>% less than 0.02 in.</u>	<u>% 0.02 to 0.1 in.</u>	<u>% greater than 0.1 in.</u>
2	600	1.5	3.5	95.0
3	600	1.3	1.7	97.0
7	600	1.1	1.5	97.4
8	600	0.9	1.4	97.7
9	600	1.7	1.4	96.9
10	600	18.6	36.6	44.8
16	600	1.9	5.3	92.8
4	100	14.5	10.0	75.5
5	100	16.9	36.2	46.8
6	100	23.1	23.6	53.3
11	100	15.0	40.7	44.3
12	100	26.0	38.7	35.3
13	100	16.3	37.5	46.2
14	100	18.3	30.7	51.0
17	100	17.9	29.7	52.4

* BNWL-1003, 68-4.

As can be seen in the above tabulation, except for Sample 10 the correlation is excellent. Those samples stored at 600 °C for 8 weeks break into fewer and much larger particles upon fracture than samples stored at 100 °C. This is probably due to the increased crystallinity observed for the samples stored at 600 °C.

There was not enough sample to make a test coupon for Sample 15.

PRODUCT MEASUREMENTS, TESTING AND STORAGE

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

At present two WSEP storage pots have been transferred into the SSETF for evaluation. However, the full utilization of SSETF has been hampered by radiation leakage problems with cubicle shielding plugs. The problem is presently being corrected and should not impair the environmental test pod storage program.

Characterization of WSEP Solidified Waste - R. J. Thompson

Numerous measurements of WSEP storage pots were taken during the quarter along with usual characterization operations.

A. Pot calorimetry measurements

1. PG-4 on 1/10/69, 2.0 kW
2. PG-5 on 1/3/69, 4.4 kW
3. SS-8 on 2/12/69, 7.4 kW
4. SS-4 on 2/13/69, 3.5 kW
5. PG-6 on 2/20/69, 4.8 kW
6. SS-9 on 4/16/69, 6.9 kW

With each pot calorimetry value an "in water" temperature profile was obtained for thermal conductivity calculations.

- B. After fill dimensions were taken on PG-4 and -6, and SS-3, -4, -5, -6, -8, and -9 pots to check for pot warpage and out-of-roundness. No significant dimensional changes were observed as compared with prefill dimensions.
- C. Pot welding was completed for Runs SS-8 and -9, and leak tests on SS-8 were completed.
- D. Temperature profiles with the pot hanging in air were taken on SS-4, -5, -6, -8, and -9 for data to be used in thermal conductivity calculations.
- E. Radiation profile scans were performed on several pots. The following are the maximum readings obtained:
 - 1. SS-3 - 1×10^4 r/hr (max), 3/3/69
 - 2. SS-4 - 2×10^4 r/hr (max), 2/26/69
 - 3. SS-6 - 3.6×10^4 r/hr (max), 3/4/69
 - 4. SS-8 - 2.8×10^4 r/hr (max), 4/4/69
 - 5. SS-9 - 2.6×10^4 r/hr (max), 4/1/69

Some difficulties have been encountered in leak testing spray solidifier pots. Continuous pumping for 32 hr on the SS-5 pot failed to reduce the pressure to values less than 100 microns where leak tests can be performed. There is some concern that the spray solidifier product is out-gassing. Study of this problem will continue.

6. HEAVY SECTION STEEL TECHNOLOGY PROGRAM

IRRADIATION DAMAGE TO HEAVY SECTION VESSEL STEELS -

C. L. Hellerich, C. W. Hunter, and J. A. Williams

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.

DCB Geometry Evaluation

Studies were conducted to evaluate the effect of specimen side groove geometry on the response of the DCB specimen. The main objective was to determine if the round side grooves would guide the crack along the specimen length, since round side grooves would be less objectional for K_{Ic} testing in light of the uncertainties associated with the three dimensional stress at the crack tip in a V-side grooved specimen. Four 2-in. specimens were machined from A533 steel and tested at -184 °F. This temperature is believed to be low enough to ensure a valid K_{Ic} evaluation as indicated by previous DCB studies. The specimens were identical except for the side groove geometry. Two specimens were machined with 60° V-notch side grooves, one with a net thickness to gross thickness ratio of $B_n/B = 0.35$ and the other with $B_n/B = 0.5$. The remaining two specimens were machined with

round side grooves, one with a side groove radius of 0.625 in. and $B_n/B = 0.35$ and the other with a side groove radius of 0.500 in. and $B_n/B = 0.50$.

The specimens were initially cracked from the machined notch at -320°F . The specimens were clamped 7 in. behind the pins to aid in the arrest of the initial running crack. Subsequent K_{I_C} testing was performed in the standard manner with the crack being reinitiated from an arrested crack in every case. In all cases, the initiation crack veered and arrested out of the central plane. In general, the cracks in the specimens with the V-notch side grooves ran straighter and longer. Because of the short crack length at which the specimen arms broke off, the two round side grooved specimens were remachined and tested. Similar results were obtained.

Because the crack ran out of plane in all tests, it was difficult to assess the significance of the data obtained in terms of calculated K_{I_C} values. The asymmetry produced by the out-of-plane crack essentially voids any calculation of K_{I_C} by the DCB equations. The K_{I_C} values could be approximated by correcting these equations for the out-of-plane location of the crack front. However, the only reasonable correction that could be made would be for the change in crack front thickness. Any other corrections would be highly speculative. Hence, any calculated results would be subject to criticism. If any conclusion can be made it would appear that round side grooves do not exhibit equal ability to the V-notched side grooves in guiding the crack along the midplane of the specimen.

Specimen Irradiations

An irradiation assembly to conduct 550°F irradiations on 1T compact K_{I_C} fracture specimens and tensile specimens has

been completed. The assembly is scheduled for insertion into the ETR during February. Discharge of the first specimens at a goal fluence of 1×10^{19} nvt (>1 MeV) is scheduled for April, 1969.

The irradiation assembly was designed for operation in the M-3 pressurized water loop of the Experimental Test Reactor (ETR). Nominal loop operating conditions are 500 °F inlet water temperature at a pressure of 2000 psi.

The irradiation assembly is shown in Figure 6.1 with the exception of the thermocouple lead tube and thermocouples. The assembly essentially consists of an outer tube, a specimen frame which holds the fracture specimen assemblies and the tensile specimen tubes, and a quick disconnect which locks the specimen frame into the outer tube. As designed, the assembly permits the removal, translation, and rotation of specimens during reactor outages. In Figure 6.2 the front specimen brace is shown open with an assembly of compact K_{IC} specimens removed. Three assemblies of four specimens are housed by the frame. Tensile specimen tubes each hold seven tensile specimens. The cycle flux monitor tube is inserted into one of the six tensile specimen tubes and is replaced during each reactor outage.

An exploded fracture specimen assembly is shown in Figure 6.3. The specimens are held in brackets by a pin-snap ring assembly. Pins are slotted for cutting the snap ring in-cell after irradiation. Specimen flux monitors are weld sealed into the set screw tube assembly shown in the foreground. These are screwed into the pin, and the assembly is installed to hold the specimen in the brackets. Though not shown in Figure 6.3, a flux monitor tube 1/8 in. in diameter and the length of the bracket is tack welded to each end of the back edge of the bracket. The monitors are removed with the assembly after goal specimen exposure.

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FIGURE 6.1. HSST Specimen Irradiation Assembly for the M-3 Hot Water Loop of the ETR. The assembly houses twelve 1T compact K_{IC} specimens and 35 tensile specimens.

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FIGURE 6.2. HSST Specimen Irradiation Assembly for the M-3 Hot Water Loop of the ETR but with the Front Specimen Brace in the Open Position. Tensile specimen, tensile specimen tube and cycle flux monitor are shown in foreground. One section of compact K_{IC} specimens has been removed.

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FIGURE 6.3. Fracture Specimen Assembly of the M-3 Irradiation Assembly. Each assembly of four specimens can be translated or rotated from reactor cycle to reactor cycle to smooth specimen fluence gradients.

The temperature of the specimen in this experimental assembly is controlled by the loop operating conditions of inlet water pressure, inlet temperature, and flow rate in combination with the gamma heating produced within the specimen. The maximum temperature calculated for the compact K_{I_C} fracture specimens is 553 °F with a surface temperature 520 °F. Thermocouples will monitor the temperature of the center of the specimens at five locations along the length of the capsule. The resultant tensile specimen irradiation temperature in this assembly will be slightly lower than the compact K_{I_C} specimens. Tensile specimen temperature is expected to be about 504 °F.

Flux monitors at various locations in the capsule previously described, are Fe and Al-Co correlation monitors in accordance with those recommended by ASTM E 184-62.

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