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NUCLEAR SAFETY QUARTERLY REPORT
MAY, JUNE, JULY 1969
FOR
USAEC DIVISION OF REACTOR
DEVELOPMENT AND TECHNOLOGY

September 1969

AEC RESEARCH &
DEVELOPMENT REPORT

ROUTE TO	DATE
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UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

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BNWL-1187

UC-41, Health and Safety

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By

The Staff of Battelle-Northwest

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PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

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NUCLEAR SAFETY BRANCH
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FOREWORD

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

PREVIOUS REPORTS IN THIS SERIES

BNWL-433	January, February, March	1967
BNWL-537	April, May, June	1967
BNWL-754	July, August, September, October	1967
BNWL-816	November, December, 1967, January	1968
BNWL-885	February, March, April	1968
BNWL-894	May, June, July	1968
BNWL-926	August, September, October	1968
BNWL-1009	November, December, 1968, January	1969
BNWL-1084	February, March, April	1969

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1. SUMMARY

ENGINEERING SAFETY SYSTEMS STUDIES

Containment Systems Experiment (CSE)

One large-scale containment spray test was performed during the quarter. In addition, the results from two large-scale runs conducted last quarter are discussed in this report. Two containment spray tests utilizing long term recirculation through the spray nozzles of the spray solution were conducted. One spray test used the basic borate solution, and the other the basic borate plus sodium thiosulfate solution. Spraying was started just before aerosol release, and recirculation of spray solutions continued for 1100 min following the introduction of fresh spray solution into the containment vessel. The test results confirm the elemental iodine removal behavior expected from earlier tests utilizing short spraying periods. The thiosulfate spray solution was more effective than the basic borate solution against both elemental iodine and methyl iodide. For example, the 2-hour dose reduction factor approached 400 for the thiosulfate spray as against about 40 for the basic borate spray solution. Methyl iodide removal half-times were about 720 min for the basic borate solution and about 80 min for the thiosulfate solution.

Results from a run involving natural pressure and temperature decay conducted after aerosol release into a steam-air containment atmosphere, are compared with those of an earlier

similar run conducted under steady state containment atmosphere conditions. The very similar removal of elemental iodine during both runs confirmed predictions of a theoretical removal model based on a natural convection heat transfer analogy. The removal half-time for elemental iodine early in the run was 14.1 min during steady state versus 16.0 min in the pressure decay run. Evidence obtained from the behavior of the cesium particulate aerosol confirmed gravity settling to be a major factor for cesium removal and diffusiophoresis to be of rather minor importance.

Correlation of the removal of methyl iodide by sprays with theoretical models was completed. The results of all CSE spray tests were consistent with a theoretical model which assumes perfectly mixed spray drops and a laminar wall film for the liquid impinging on the containment vessel walls. This model does not account for the decreased transfer rate observed in one run which included a surfactant. The most recent spray run included an additional experiment in which no spray drops were present in containment but the basic borate thiosulfate solution was directed along the wall of the containment vessel so that only a wall film of solution was effective against the methyl iodide. The removal half-time was about 100 min compared with about 80 min for the same solution in sprays. The theoretical model prediction of a less effective removal by the wall film is accounted for by the observation that the wall film in run A-12 was highly turbulent rather than laminar as assumed in the model.

A recirculating air cleaning system has been installed in the CSE containment vessel in preparation for tests of the effectiveness of recirculating filter systems in decontaminating the containment atmosphere to be conducted next quarter.

In a cooperative effort with Battelle-Columbus, an intermediate scale evaluation was made of the effectiveness of a

reactive surface coating developed by Battelle-Columbus for rapid reaction with methyl iodide. The experiments are described and results are summarized. The work will be reported on by Battelle-Columbus.

Results from seven blowdown runs with the CSE reactor simulator vessel are reported. These runs were from the middle blowdown nozzle located just below the dummy core plate containing multiple orifice holes. Measured liquid levels during the blowdown transient are compared with predictions of the FLASH2 code. Results show formation of a separate vapor dome below the plate for runs with a 4-in. break size, but not for those of 1 or 2-in. break sizes. FLASH2 did not account for counter-current bubble rise against the liquid mixture. Hence, FLASH predictions using a two-volume vessel were poor for the smaller runs. It has been possible to establish that, from experimental results for blowdowns giving a mixture velocity downwards through the plate greater than 4.6 ft/sec, no bubble rise through the plate occurs. These results illustrate a deficiency of the FLASH code and provide the basis for incorporating improvements.

Transient forces on the core plate, measured during this series of blowdowns, confirm results reported last quarter that the equivalent differential pressure derived from the strain bolts supporting the core plate are smaller by a factor of 3 to 4 than the differential pressures predicted by the WATERHAMMER code for the experiment.

Particle Transport to Surfaces by Condensing Steam

The theoretical analysis of aerosol transport to a vertical surface upon which steam is condensing was completed and the work is being documented in a topical report. The analysis was modified in the later stages to account for thermophoresis effects which resulted in a nominal deposition rate change.

PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIESCrack Detection in Pressure Piping by Acoustic Emission

Investigation of wedge mountings for coupling the shear component of acoustic emission signals from a plate surface to a sensor has been extended to the study of heavy section 4-in. thick steel plate. Work to date, using PZT-5 sensors, shows improved sensitivity for the wedge mount versus surface mount up to 1.5 MHz, but the signal front obtained through the current wedges does not have good definition.

Maximum acoustic emission detection range in 4-in. thick steel using various sensors is being evaluated. Thickness-poled, surface-mounted PZT-5 sensors appear to be limited to 4 to 5 ft for reliable detection. However, edge-mounted PZT-5 shear sensors receive well up to at least 8 ft (maximum dimension of the specimen). Detection frequencies used were 500 ■ 700 kHz.

A simple method for obtaining relative calibration of sensor response characteristics has been developed using a small air abrasive unit to provide a broad band excitation source.

Lithium niobate used in the shear mode shows good sensitivity to acoustic emission and offers definite potential for use in a high temperature sensor. This material is a piezoelectric with a service temperature range >1500 °F.

PZT-5A piezoelectric crystals exposed to a total of 4.9×10^8 R of gamma radiation showed no detectable effect.

Tuned preamplifiers of the type used by BNW for acoustic emission detection have been tested for resistance to temperature damage. Results show them capable of withstanding 185 °F for an extended period. Long term service at 200 °F is attainable by using special high temperature wire and would meet anticipated temperature requirements for continuous surveillance of thermal power reactors.

A high pass filter incorporating 36 db per octave attenuation has been developed and tested. This filter will enhance ability to detect low level acoustic emission signals in the presence of lower frequency background noise.

On the basis of total engineering information, sections of process piping in local reactors monitored for fatigue crack growth by acoustic emission detection are to be replaced. Arrangements have been made to salvage for examination those sections producing limited acoustic emission indications of crack growth. This examination will contribute to establishing a criterion for evaluating acoustic emission indications.

Increasing the monitor frequency from >200 kHz to >600kHz on the acoustic emission monitor applied to the Containment System Experiment simulator vessel was found to reduce noise interference without degrading the data.

FIXATION OF RADIOACTIVE RESIDUES

To date, 23 radioactive demonstrations in WSEP have been completed with simulated wastes containing approximately 32 MCi of mixed radionuclides converted to solids.

During the report period, approximately 2.6 MCi of radioactivity were processed in the tenth spray solidification run, and approximately 2.7 MCi in the seventh phosphate glass run. Spray solidification run SS-10 was made using a simulated Purex Waste 4m (PW-4m) and was the last in the first series of spray solidification runs. Run SS-10 completed that phase of the Waste Solidification Demonstration program wherein the basic engineering feasibility of all three solidification processes has been established. Although the spray solidification system is certainly not optimized, the performance data have shown that major processing factors can be adequately controlled.

Phosphate glass Run PG-7 was made using a simulated Purex Waste 2 (PW-2) and is the first in the final series of phosphate glass demonstrations. The PW-2 glass effective thermal conductivity of $0.9 \text{ W/(m}^2\text{)} \text{ (}^\circ\text{C/m)}$ during air storage increased to $1.85 \text{ W/(m}^2\text{)} \text{ (}^\circ\text{C/m)}$ when most of the glass was molten during the pot filling operation in the receiver furnace.

Development spray solidification runs indicated that greater than 95% of the ruthenium can be retained in the calcine from PW-4m feeds over a range of acidities if the phosphate flux is not added to the feed.

Three Distillation Demonstration Unit (DDU) runs have been made using sulfate containing condensate from phosphate glass solidification Run PG-4. At operating pressures of 30 mm Hg and lower, the sulfuric acid fraction can be distilled with a low nitric acid content and with a ruthenium content corresponding to a DF of greater than 200, a value large enough to permit incorporation of the sulfuric acid in asphalt for permanent storage. Vacuum distillation of the melter condensate has been shown on engineering scale equipment to be a viable concept for decontamination of the sulfuric acid fraction.

2. ENGINEERED SAFETY SYSTEMS STUDIES

CONTAINMENT SYSTEMS EXPERIMENT - G. J. Rogers

Fission Product Transport Studies - R. K. Hilliard

One large-scale containment spray experiment, Run A-12, was performed during this report period. The results of this experiment, together with those of two experiments performed last quarter, are discussed. Several ancillary experiments were performed during the large-scale tests, and preliminary results of two of these, fission product leakage and methyl iodide removal by reactive spray and wall films, are reported. Small-scale tests were made to evaluate the effectiveness of a reactive paint in removing iodine and methyl iodide from containment atmospheres. Equipment was installed in the CSE vessel in preparation for air cleaning experiments to be conducted next quarter.

Removal of Iodine by Recirculating Sprays - A. K. Postma

Reactor containment sprays are designed to start before significant quantities of fission products are released from overheated fuel. Hence, under design basis accident conditions, fuel materials and fission products released into the containment vessel will be scrubbed by sprays during the release period. In earlier CSE spray experiments, relatively short spray periods were used to permit assessment of the important parameters influencing spray washout. In Runs A-10 and A-12, the sprays were turned on shortly before injection of elemental iodine and cesium into the containment vessel. This operating sequence more nearly duplicates the conditions expected in postulated reactor accidents.

Run A-10 used a basic sodium borate solution made by adding excess sodium hydroxide to boric acid. The final pH was 9.4. Spraying was started 2 min before time zero, defined as the beginning of injection of elemental iodine. Fresh

solution at room temperature was sprayed at 50 gal/min for 42 min. Sprays were then shut-off for 10 min while valving was changed to permit recirculation of the liquid collected in the vessel sumps to the spray nozzles. Recirculation was continued for about 1100 min.

The gas phase concentrations versus time for elemental iodine, particulate iodine, and methyl iodide as determined from Maypack gas samples are shown in Figure 2.1. Elemental iodine is defined as the species adsorbed on the silver components of the sampler, particulate iodine as the species trapped by the inlet glass fiber filter, and methyl iodide as the species trapped on the charcoal bed of the sampler. The standard deviation of concentrations indicated by simultaneous gas samples is shown as a vertical bar associated with the plotted mean concentration.

All iodine forms increased in concentration during the injection period, and then decreased in concentration with time after injection was completed. Particulate associated iodine decreased rapidly in concentration to a value of about 1% of the elemental iodine concentration. Methyl iodide was removed slowly, showing a concentration half-life of 720 min. Elemental iodine decreased rapidly in concentration for several minutes following the end of the injection period and then decreased at a slower rate for the remainder of the spray period. An elemental iodine removal half-time of about 200 min was observed for the 1100-min recirculating spray period.

The results of Run A-10 are in qualitative agreement with earlier CSE spray experiments employing short spray periods. The most important quantitative difference is that the reduction in elemental iodine washout rate in Run A-10 occurred at a higher gas phase concentration than in earlier tests. Results from the earlier tests would have suggested decrease by an

FIGURE 2.2 *Spray Washout of Iodine By Basic Sodium Borate Solution*

additional factor of 10 in the elemental iodine before beginning of the slower washout period. One explanation for this behavior was the formation, subsequent to injection of an iodine species producing over the spray solution an appreciable vapor pressure adsorbed on the silver components of the Maypack sampler. We have no information at present to account for the different behavior.

This slower than anticipated removal of iodine by the basic borate spray may not be of great practical importance because the 2-hr dose reduction factor estimated from the observed gas concentration and the iodine injected into the vessel was about 40. Calculations⁽¹⁾ indicate that, for the TID-14844 fission product release assumptions, the noble gas doses become controlling for iodine dose reduction factors greater than 13. If the elemental iodine behavior had been as expected from the earlier runs, the 2-hr dose reduction factor would have been about 40. This estimate is based on a removal rate constant of 0.37 min^{-1} (measured in Run A-6) and an equilibrium partition coefficient of 3×10^4 (measured after 260 min in Run A-8). Nevertheless, the dose reduction factor obtained from Run A-10 is in good agreement with results expected on the basis of previous tests. The relatively slower washout observed in Run A-10 was off-set by a peak gas phase concentration lower than predicted, and these factors combined to give an average concentration in agreement with that expected from earlier CSE results.

In Run A-12, sodium thiosulfate (1 wt%) was added to the spray solution. Also methyl iodide was injected about 24 hr prior to the start of sprays to permit study of its containment behavior. The physical conditions and operating sequence was otherwise similar to Run A-10. The concentrations of airborne iodine species in Run A-12 are shown in Figure 2.2.

FIGURE 2.2 *Spray Washout of Iodine by Basic Sodium Thiosulfate Solution*

Methyl iodide was removed about 10 times faster by the thiosulfate spray in Run A-12 than by the basic borate solution used in Run A-10. This enhanced removal rate for methyl iodide is expected on the basis of the reaction rate of methyl iodide with the thiosulfate solution. The observed removal rate is compared to theoretical predictions in a subsequent section of this quarterly report.

The sum of the concentrations of elemental iodine and particulate associated iodine is also shown in Figure 2.2. These two components are not shown separately because capture of an appreciable fraction of the elemental iodine by the fiberglass filter wetted by a small amount of thiosulfate solution is anticipated. The sum of elemental and particulate forms is rapidly diminished by the operation of the thiosulfate spray. The 2-hr dose reduction factor for elemental and particulate iodine forms is estimated to be about 400, an appreciable increase over that observed for the basic borate solution. For part of the recirculating spray period, the elemental and particulate iodine curve is parallel to the methyl iodide curve. This result may be due to absorption of a small amount of methyl iodide by the fiberglass filter. All iodine forms decrease to a very low level before the gas phase concentration appears to approach a constant value.

Natural Transport During Pressure Decay - R. K. Hilliard

Run A-11 was a natural response experiment, i.e., no engineered safety features were operated and fission product behavior was observed during natural temperature and pressure decay. The run differed from previous CSE natural response experiments in that the earlier tests (Runs A-1, A-2 and A-5) were performed at steady state thermal conditions, with the replacement of heat losses by addition of steam. In Run A-11, the containment vessel and atmosphere were at thermal

equilibrium at the time the fission product simulant materials were released, but the steam feed was terminated as soon as the aerosol release was completed. Heat transfer through the insulated walls of the CSE vessel caused the temperature and pressure to decline continuously throughout the ensuing 48-hr test period. Table 2.1 lists the experimental conditions, along with those for Run A-5, a steady state atmosphere test.

TABLE 2.1. *Summary of Test Conditions, Runs A-5 and A-11*

	<u>Run A-5</u>	<u>Run A-11</u>
Type of Test	Natural response, isothermal, isobaric	Natural response, temperature & pressure decay
Vessel	Main CSE vessel plus open drywell. Wetwells sealed off.	Same as A-5
Previous exposure to steam, hr	586	1500
Avg. temp., main room, °F	253 ± 3	252 ± 2 ^(a)
Avg. temp., middle room, °F	233	218
Avg. temp., lower room, °F	210	160
Avg. total pressure, psia	46.5 ± 0.6	49.1 ^(a)
Avg. steam makeup, lb/hr	295 ± 20	0
Condensing steam flux on main vessel walls, lb/hr-ft ²	0.032 ± 0.006	0.046 ^(a)
Aerosol released duration, min.	10	10
Calculated initial concentration in main room, mg/m ³ (b)		
Total iodine	140	165
Methyl iodide	1.6	1.6
Cesium	1.8	8.4
Uranium-Zircaloy	~3	~3

- a. *Initial condition only. Temperature and pressure decreased with time after aerosol release was completed.*
- b. *Based on the total mass of each material delivered to the containment vessel divided by the net free volume.*

FIGURE 2.5 *Steam and Heat Flux, Run A-11*

FIGURE 2.6 Concentration of Various Iodine Forms in Containment Atmosphere, Run A-11

FIGURE 2.7 Comparison of Elemental Iodine Concentration in Gas Space for Constant Temperature and Pressure Decay Conditions

for temperature and pressure decay (Run A-11). The two curves are very similar and agree closely with predictions based on natural convection heat transfer analogy. (2)

The behavior of particulate materials is dependent on condensing steam flux and atmospheric conditions, as expected. The concentration of cesium is plotted against time for Runs A-5 and A-11 in Figure 2.8. Identical concentration half lives of 42 min were measured in both tests initially. However, the half life in the pressure decay test increased to about 500 min after about 2 hr, while that in the isothermal test increased only to 100 min. This behavior can be explained qualitatively by the fact that aerosol particles are transferred from the gas space to surfaces by three major mechanisms: (1) gravity settling to horizontal surfaces, (2) turbulent diffusion across gas boundary layers at all surfaces, and (3) diffusiophoresis to steam condensation surfaces. The rates of transfer by the first two mechanisms are dependent on particle size, with more rapid transfer for larger particles. The rate by diffusiophoresis is essentially independent of particle size but is directly proportional to steam condensation flux. If diffusiophoresis were the only mechanism responsible for removal from the gas phase, the concentration half life of cesium in Run A-5 would have been invariant with time at a value of 740 min, according to Equation (1).

$$t_{1/2} \text{diffusiophoresis} = \frac{(0.693) (18) P V}{n_s R T A} \quad (1)$$

where P = total pressure psia
 V = gas volume, ftf
 n_s = steam condensation flux, lb/hr-ft²
 R = gas constant = 10.73 psia ft³/lb-mole/°R
 T = gas temperature, °R
 A = heat transfer area, ft²

FIGURE 2.8 Comparison of Cesium Concentration in Gas Space for Constant Temperature and Pressure Decay Conditions

Equation (1) is based on the simplifying assumption that the aerosol particles move with the gas bulk motion resulting from steam condensation. A more detailed theoretical treatment of the diffusiophoretic velocity for CSE conditions⁽³⁾ indicates that this assumption would lead to inaccuracies smaller than about 10%.

The measured concentration half-life, much less than the calculated value for diffusiophoresis, indicated that diffusiophoresis played a minor role in transferring the cesium from the gas. Integration of the wall trough flow and concentration over the duration of the experiment showed that only 6% of the cesium reached the vertical walls. Thus it appears that gravity settling was the major mechanism in Run A-5, with a mean aerosol particle diameter of about 10 μ initially and 8 μ after the initial 2 hr.

For Run A-11, a similar conclusion is reached. Only 5.8% of the cesium reached the vertical walls. The gas concentration behavior was consistent with gravity settling of 10- μ diam aerosol particles initially, then with 4- μ diam particles. Efforts are continuing to differentiate between the three removal mechanisms.

Methyl Iodide Removal by Sprays - A. K. Postma

Methyl iodide is one of the chemical forms of radioiodine expected to exist in the containment vessel atmosphere following postulated fuel overheating accidents in water-cooled nuclear reactors.

Spray systems incorporated into reactor containment systems provide a means for rapidly removing airborne elemental iodine, and the effectiveness of water sprays against elemental iodine has been demonstrated on a large scale in the CSE vessel.⁽³⁻⁵⁾ For methyl iodide, however, the spray washout rate is much slower. The magnitude of the washout rate depends on the rate at which methyl iodide is destroyed by chemical

reaction within the spray solution. The spray washout of methyl iodide has been studied in CSE experiments for basic sodium borate solutions, and for basic sodium borate solutions containing sodium thiosulfate.

Methyl Iodide Absorption by Sodium Borate Solutions. One solution proposed for use in containment spray systems will contain 3000 ppm boron as boric acid, neutralized with excess sodium hydroxide to give a final pH of about 9.5. This solution has been used in many CSE spray tests, and effective removal of elemental iodine has been demonstrated. However, this solution is nominally unreactive with methyl iodide, and hence the spray removal rate is slow for iodine bound chemically as methyl iodide. The reaction rate of methyl iodide with the basic borate solution would be expected to be governed by the reaction rate of methyl iodide with the water molecule. (6)

The removal of methyl iodide by recirculating spray of borate solutions is summarized in Table 2.2. In all runs except Run A-10, the methyl iodide removal rate was too small to be determined accurately. In Run A-10, the spray period was long enough to cause an appreciable decrease in the methyl iodide concentration, and the removal half-time indicated was 720 min. Similar results for once-through sprays, allowing for intercompartmental mixing, were obtained.

It is concluded from these results that, for sprays of boric acid basic solution, the methyl iodide dose reduction factor for the initial 2-hr period following an accident would be about unity. The observed removal rate is rapid compared to the rate of radioactive decay, and hence dose reduction factors calculated for long time periods would be significantly greater than unity.

TABLE 2.2. *Methyl Iodide Removal By Recirculating Water Sprays In CSE*

<u>CSE Run</u>	<u>Spray Duration, min</u>	<u>Spray Solution</u>	<u>Drop Diameter, *</u> <u>μ</u>	<u>Flow Rate, gal/min</u>	<u>CH₃I Removal Half-Time, min</u>
A-4	45	a	1210	48.8	> 500
A-6	60	b	1210	49.0	> 500
A-7	60	c	1210	49.0	> 500
A-8	60	b	770	50.5	> 500
A-10	1090	b	1210	49.0	720
a - 525 ppm B as H ₃ BO ₃ in NaOH, pH 9.5					
b - 3000 ppm B as H ₃ BO ₃ in NaOH, pH 9.5					
c - 3000 ppm B as H ₃ BO ₃ , pH 5					

* *Mass median diameter*

Methyl Iodide Absorption by Sodium Thiosulfate Solution.

Sodium thiosulfate in basic solution is presently considered for use as a reactive additive in spray systems of some commercial nuclear plants. It is known to react very rapidly with elemental iodine, and relatively rapidly with methyl iodide. A number of spray tests have been carried out in the CSE system in which 1 wt% sodium thiosulfate was added to the basic borate solutions. The removal of methyl iodide by the thiosulfate sprays is summarized in Table 2.3.

Comparison of the results presented in Table 2.2 and Table 2.3 shows that the thiosulfate increases the washout rate by a factor of 5 or more. The enhanced rate is, however, only a few percent of the rate calculated for absorption limited by gas phase mass transfer resistance.

TABLE 2.3. *Removal Of Methyl Iodide By
Thiosulfate Sprays in CSE*

<u>CSE Run</u>	<u>Spray Dura- tion, min</u>	<u>Spray Solution</u>	<u>Drop Diameter," M</u>	<u>Flow Rate, gal/min</u>	<u>Average Tempera- ture, °F</u>	<u>CH₃I Removal Half-Time, min</u>
A-7	50	a	1210	49.0	210	130
A-8	50	a	770	50.5	212	140
A-10	240	b	1210	50.0	246	85
A-10	240	c	1210	50.0	246	155
A-12	1270	b	1210	50.0	245	80
A -	120	b	wall	50.0	246	100
a - fresh solution, 3000 ppm B, 1 wt% Na ₂ S ₂ O ₃ , pH 9.2						
b - recirculated solution, 3000 ppm B, 1 wt% Na ₂ S ₂ O ₃ , pH 9.2						
c - Same as b, but with 0.6 wt% PEI* surfactant						

* *Mass median diameter*

In Runs A-7 and A-8, the spray periods were short compared to the removal half-time. Hence, the indicated half-times would not be expected to be highly precise for these runs.

In Run A-10, two spray periods were appended at the end of the run. For the first period, 1 wt% thiosulfate was added to the basic borate solution recirculated throughout the main part of this run. For the second spray period, a surfactant, PEI-1000,* was added to the spray solution to determine its effect on methyl iodide absorption. It appears that the surfactant decreased the absorption rate. The decreased mass transfer rate was probably caused by interfacial resistance, or by reduced mixing near the gas-liquid interface.

* *Manufactured by Dow Chemical Company, Midland, Michigan.*

In Run A-12, methyl iodide absorption by a long term recirculating thiosulfate spray, and by a wetted wall was measured. A washout half-time of about 80 min was observed for the long term spray. This removal rate continued until the methyl iodide concentration decreased to the detection limit of the Maypack samplers. For the wetted wall experiment, the spray liquid was ducted to 1/4 in. tubes aimed tangentially against the outer wall of the vessel. About 50% of the wall surface between the spray header and the floor of the main room was wet by the spray solution. No drops were generated during this part of the run. The washout half-time measured for the wetted wall test was about 100 min, as compared to 80 min for the regular spray. Considering the relatively small area exposed during the wetted wall test, this experiment demonstrates that wall absorption is highly significant in methyl iodide absorption by thiosulfate sprays.

Comparison of Observed Methyl Iodide Removal With Theoretical Predictions. Methyl iodide absorption was predicted on the basis of a model which considered the drops to be perfectly mixed and the wall film to be laminar. The wall film thickness was estimated from the measured wall flow rate on the outer shell of the containment vessel. The surface area exposed by wall film liquid was assumed to equal all surface area exposed to the containment atmosphere in the main room of the CSE vessel. This total surface area, 6140 ft^2 , is about twice that of the surface area exposed by the vessel shell in the main room. This model would be expected to overestimate the relative influence of the falling drops as compared to the wall film.

For well mixed drops, the methyl iodide removal half-time is calculated to be:

$$t_{1/2} \text{ (drops)} = \frac{0.693 V}{LH (1 + kt_e)} \quad (2)$$

where $t_{1/2}$ = gas phase removal half-time for drops,
 V = volume of the gas phase,
 L = volumetric flow rate of drops,
 H = gas-liquid partition coefficient for CH_3I ,
 K = first order reaction rate constant for CH_3I
in liquid,
 t_e = drop exposure time.

The drop exposure time was calculated on the basis of the drops falling at terminal velocity for the average fall height of 38 ft.

For the wall film, the methyl iodide removal half-time is calculated to be

$$t_{1/2} \text{ (wall)} = \frac{0.693 V}{AH \sqrt{kD} \tanh\left(\sqrt{\frac{k}{D}} \delta\right)} \quad (3)$$

where $t_{1/2} \text{ (wall)}$ = gas phase removal half-time for wall film.
 A = surface area of film,
 D = diffusivity of CH_3I in liquid film,
 δ = thickness of the wall film.

Equation (3) applies for a stagnant film of thickness δ . The diffusivity of methyl iodide in water, D , was estimated from a correlation published by Wilke and Chang. (7)

The wall liquid film thickness was predicted from theory on the basis of laminar flow on a smooth vertical surface with no shear at the gas-liquid interface:

$$\delta = \left(\frac{3 \nu \Gamma}{g} \right)^{1/3} \quad (4)$$

where δ = thickness of wall film,
 ν = kinematic viscosity of liquid,
 Γ = liquid film flow rate per unit length,
 g = acceleration due to gravity.

The partition coefficient for methyl iodide, H , was measured as a part of this study. A complete description of the methods used and the results obtained are being prepared for publication. The reaction rate constant, k , was predicted from measurements reviewed in Reference (6). The input data used in evaluating the theoretical predictions are summarized in Table 2.4.

TABLE 2.4. *Data Used in Model for CH_3I Washout*

Run	k (sec^{-1})	Γ ($\text{cm}^5/\text{sec cm}$)	v (cm^2/sec)	D (cm^2/sec)	t_e (drops) (sec)	H
A-7	1.3	0.045	2.4×10^{-5}	5.6×10^{-5}	3.2	1.14
A-8	1.35	0.079	2.4×10^{-3}	5.6×10^{-5}	4.8	1.14
A-10	4.5	0.061	2.0×10^{-3}	7.2×10^{-5}	3.2	1.08
A-10	4.5	0.061	2.0×10^{-3}	7.2×10^{-5}	3.2	1.08
A-12	4.4	0.061	2.0×10^{-3}	7.1×10^{-5}	3.2	1.08
A-12	4.5	2.63	2.0×10^{-3}	7.2×10^{-5}	(wall)	1.08

The predicted removal half-time due to washout by falling drops and by wall film liquid are compared to the measured removal half-time in Table 2.5.

TABLE 2.5. *Comparison Of Predicted And Measured Methyl Iodide Washout Rates*

CSE Run	Wall Flow, gal/min	Predicted $t_{1/2}$ For Wall Film, min	Predicted $t_{1/2}$ For Drops, min	Predicted Overall $t_{1/2}$, min	Measured $t_{1/2}$, min
A-7	1.7	200	520	145	130
A-8	3.0	180	340	120	140
A-10	2.3	83	180	57	85
A-10	2.3	83	180	57	155
A-12	2.3	83	185	57	80
A-12	50.0	382	-	382	100

The measured and predicted removal half-times are in good agreement, except for the second spray periods of Runs A-10 and A-12. In the second period of Run A-10, the transfer rate was decreased by the surfactant, the presence of which was neglected by the theory. In Run A-12, the highly turbulent wall film increased the washout over that for a laminar film. This enhancement, evident in Table 2.5, indicates the measured washout rate to be about four times faster than predicted for a stagnant film.

Large Air Cleaning System Experiments - C. E. Linderoth
and J. D. McCormack

A series of experiments will begin next quarter to evaluate the effectiveness of internal recirculation air cleaning loops for decontaminating postaccident containment atmospheres. A schematic drawing of the loop as it has been installed in the CSE vessel is shown in Figure 2.9. The loop is assembled of modular units, each to contain a commercial 2 ft by 2 ft cleaning component. Provision is made for measuring differential pressure, temperature, and fission product concentration between each component in the loop. In addition, the containment vessel atmosphere will be monitored for fission product simulant concentration, temperature, and pressure as in all previous CSE experiments.

The first experiment to be performed with the recirculation loop will be with room temperature and pressure air at 100% relative humidity. Following tests will be with pressurized steam-air atmospheres.

Test of a Methyl Iodide Absorptive Surface Coating
R. H. Bond

Battelle Memorial Institute, Columbus Laboratories,, has developed surface coatings capable of reacting rapidly with methyl iodide.⁽⁸⁾ Such coatings would be applied to the

FIGURE 2.9 Sketch of Air Cleaning Loop in the CSE Vessel

interior surfaces of a containment structure to rapidly reduce the methyl iodide concentration in the atmosphere to a very low level.

A BMI formulated coating (DMAM ⁽⁹⁾ formulation two coats) was tested in the ADF Laboratories in cooperation with BMI-Columbus. The coating, applied over the existing Pheno-line 302 surface of the painted aerosol tank (PAT) and air dried for two weeks, was evaluated under containment conditions to supplement BMI-Columbus evaluation of the coating applied to small test panels. Tests were run under stable equilibrium conditions of 80 °C, ambient atmospheric pressure, steam-air atmosphere with vigorous agitation of the tank atmosphere, and recirculation of condensate for agitation and mixing.

Methyl iodide and elemental iodine used for the tests were radioactively traced. Results were evaluated by gamma spectroscopy tracer techniques.

The coating was tested first with methyl iodide alone at 10 mg I/m³ initial concentration. Atmosphere and condensate samples were taken over a 24 hr period. The run was then terminated without further steaming for recovery.

Results showed atmosphere depletion by four orders of magnitude in 300 min for a half-life of 16.5 min. Approximately 50% of the injected methyl iodide was recovered in the condensate by the time the run was terminated at 24 hr. An additional 35% of injected methyl iodide was recovered as condensate when steaming was begun for the second test of the coating.

The coating was tested in a second run with essentially instantaneous (2-min duration) injection of methyl iodide (10 mg I/m³) and elemental iodine (100 mg I/m³).

Intensive atmosphere sampling was conducted to define initial conditions (2-min samples at 4-min intervals starting 1 min after completion of injection). Sampling continued for 24 hr. The tank was cooled for test-panel recovery, then was steamed to essentially complete recovery (negligible iodine concentration in recovered condensate).

Results for this second test showed a methyl iodide half-life of 16.2 min, essentially identical with the first test.

Depletion of elemental iodine from the atmosphere was very rapid. The earliest atmosphere samples showed a half-life of about 1.3 min, but initial half-life could have been as low as 0.5 min.

Tests were run in the Phenoline 302-coated tank as a basis for comparison prior to application of the BMI coating under conditions essentially the same as the BMI coating tests. Comparison showed the BMI coating to be about 33 times more effective for methyl iodide absorption than Phenoline 302 coating and that it apparently absorbed elemental iodine more rapidly initially than did Phenoline 302. However, the amount of iodine in both coatings at complete recovery was essentially identical.

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 reactor dummy core structures.

During this report period, a series of blowdown runs was made from a nozzle on the side of the vessel below a perforated plate dummy core installed in the vessel. The purpose of the series was 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.

Blowdowns were made from the middle nozzle of the CSE simulator vessel, with a nozzle extension and orifice plate as shown in Figure 2.10. The core plate divided the vessel volume to give a ratio of upper to lower volume of 2.3. Sixty-four nozzles of 1.23 in.² each were installed, and the remaining 24 holes were plugged. The flow area of this sieve plate was 0.56 ft². Blowdown was initiated using the double-rupture-disc, over-pressurization technique. The experimental arrangements for the blowdown series reported herein is described extensively in earlier quarterly reports.^(4,5) The major changes made for this series involved location of the blowdown nozzle and the insertion of double pressure probes in nozzles to obtain pressure near the centerline and at the wall of the vessel simultaneously.

The initial conditions for the blowdown experiments performed this period are summarized in Table 2.6.

FIGURE 2.10 CSE Reactor Simulator Vessel with Core Plate

TABLE 2.6. *Blowdown Run Conditions*

<u>Run No.</u>	<u>Nominal Nozzle Diam, in.</u>	<u>Press, psia</u>	<u>Temp, °F</u>	<u>Over Press, psia</u>	<u>Nominal Vessel Volume</u>	<u>Area Break Free Area Plate</u>
B-22	2	1330	500	650	Full	0.028
B-22B	2	1301	500	621	Full	0.028
B-23	1	2152	500	1472	Full	0.0064
B-25	4	1300	500	620	Full	0.115
B-26	2	1020	545	170	3/4 Full	0.028
B-27	4	1275	545	272	3/4 Full	0.115
B-27B	4	1090	545	87	3/4 Full	0.115

Comparisons of FLASH2 Predictions and Experimental Data
for Liquid Level and Vessel Pressure - A. S. Neuls

The FLASH2 code⁽¹⁰⁾ was applied to five of the CSE blowdowns performed during the period of this report, B-22B, B-23, B-25, B-26, and B-27B. Table 2.7 contains a list of parameters used in applying the code to the CSE blowdowns. Comparisons were made between predicted and experimental liquid level and pressure histories.

Figures 2.11 through 2.13 show observed liquid level histories and FLASH2 predictions. The experimental data are taken from a time domain reflectometer probe measuring, essentially, the distance from the top of the vessel to an electronic impedance discontinuity caused by the upper surface of the liquid or foam. As long as liquid remains above the dummy core plate, the probe receives no information concerning liquid level in the lower volume. When the liquid level in the upper volume is gone, the indication suddenly "collapses" to the liquid level reading in the lower volume. The time and pattern of the collapse are used as a measure of the liquid level behavior. Some of the discrepancy between the predicted curve and the observed level data can be attributed to

TABLE 2.7. FLASH2 Parameters

Run No.	No. of Control Volume	Nominal Size, in.	Break Diam, in.	Initial Vol, ft ³	Discharge Orific ^a Coefficients	Bubble ^(a) Rise Velocity, ft/sec	Quality Gradient Factor
B-22B	1	2	1.689	149	Low-Qy 0.80	2.0(b)	0.8
B-23	1	1	0.815	149	1.049	3.0	0.8
B-25	2	4	3.438	149	0.664	2.0, 2.0(b)	0.8, 0.4(a)
B-26	1	2	1.689	113.6	0.80	2.0(b)	0.8
B-26	2	2	1.689	113.6	0.80	3.0, 3.0	0.8, 0.8(a)
B-27B	2	4	3.438	113.5	0.664	2.0, 2.0(b)	0.8, 0.4(a)

(a) For two-volume cases, the first number designates the upper volume parameter, the second number the lower volume parameter.

(b) Variable bubble rise model from J. A. Redfield and J. H. Murphey, Void Fraction and Residual Water Predictions During Loss-of-Coolant, APD-T-2155, Bettis Atomic Power Laboratory, Pittsburgh, Pa., September 1966.

FIGURE 2.11 Level of Fluid Remaining in Vessel, Run B-23 MN

FIGURE 2.12 *Level of Fluid Remaining in Vessel, Run B-25 MN*

FIGURE 2.13 Level of Fluid Remaining in Vessel, Run B-22 BMN

geometric differences between the FLASH2 model, a right-circular cylinder, and the actual reactor simulator vessel, a cylinder with hemispherical ends. If total remaining volume rather than liquid level were being compared, better agreement could be obtained.

The TDR data is known to contain a temperature drift for which no corrections have been made in the figures. Corrections for this drift would serve to continue the decrease in level after the transition region (inflection point in pressure curve at which high quality blowdown begins) rather than to maintain it constant as shown for most of the runs.

Figure 2.13 illustrates the results of two FLASH2 predictions for Run B-26. The prediction made on the basis of a two-volume vessel was not as good in predicting level drop pattern as was the single-volume prediction. The major level discrepancies are primarily due to inaccuracy of the initial fluid level and its effect on the level gage calibration. The shape remains similar in spite of this inaccuracy. The more detailed two-volume approximation, which includes the sieve-plate restriction, indicated a level drop at the time the level reached the sieve plate (level collapse). The FLASH2 model in this case predicts the trapping of a steam volume under the plate since the model does not allow for counter-current bubble rise between the two volumes. The experimental data indicated little or no steam volume under the plate and, hence, there was counter-current bubble rise through the plate. Agreement of the single-volume model for B-26 was much better since it contained no plate restriction and allowed bubble rise to occur.

Further evidence indicating bubble rise through the plate for B-26 was found by comparing the pressure decay curves in Figure 2.14. The two-volume model predicted, before transition, a change in pressure decay slope that did not appear in the experimental data.

FIGURE 2.14 FLASH2 Prediction, Pressure De in Vesse

Comparisons of data and predictions for the remaining four runs indicated a counter-current bubble rise through the sieve plate for the small break diameter blowdowns (1 and 2-in.) but not for the intermediate break size (4-in.). For B-22, a full vessel **blowdown** with a 2-in. break, the single volume FLASH2 prediction showed good agreement with both the pressure curve, Figure 2.14, and the liquid level history, Figure 2.13.

For B-23, a full vessel **blowdown** with a 1-in. break, the single-volume prediction again was in good agreement with the liquid level history as shown in Figure 2.11. However, for larger (4-in. nominal) break size Runs B-25 and B-27 (Figure 2.12), the two-volume model gave superior results. In these runs, the bubble passage upward through the plate was inhibited.

The level history for B-25 (Figure 2.12) showed a level collapse near the transition from low-quality to high-quality flow, thus indicating steam volume entrapment under the plate and consequent inhibiting of bubble rise through the plate. For B-27 (Figure 2.12), a 3/4 full vessel blowdown, the level history indicates the same level collapse as seen in the B-25 data.

In an effort to define the approximate value of the through-plate liquid velocity, above which bubble rise is inhibited and below which bubbles rise freely through the plate, an investigation of the liquid level data from previous sieve plate runs was made. Results are given in Table 2.8. For those blowdowns exhibiting total collapse to approximately the nozzle level, the nominal mixture velocity through the plate was well above 4.6 **ft/sec.** For those blowdowns showing no level collapse, the nominal mixture velocity through the plate was below 4.6 **ft/sec.**

TABLE 2.8. *Nominal Mixture Velocities
Through The Sieve Plate Orifices*

<u>Run No.</u>	<u>Break Position</u>	<u>TDR Trace</u>	<u>Nominal Velocity, ft/sec</u>
B-19B	bottom	partial collapse	4.6
B-20B	bottom	total collapse	7.8
B-21	bottom	total collapse	15.9
B-22B	middle	smooth	2.6
B-23	middle	smooth	0.9
B-24	middle	total collapse	7.5
B-25	middle	total collapse	7.8
B-26	middle	smooth	3.1
B-27	middle	total collapse	8.5

In B-19B, a bottom nozzle blowdown, the liquid level (Figure 2.15) collapsed when it reached the plate but did not drop to the exit nozzle level. In all other runs indicating steam volumes under the plate, the level collapsed from the plate to about the elevation of the exit nozzle. Moreover, the slope of the level curve for B-19B did not change after the collapse through the plate. This fact indicated that the exit flow was still in the low-quality region, i.e. before transition. For all other cases exhibiting level collapse, the change in level curve slope after the level dropped indicated a change in the flow regime from low-quality to high-quality near the time when the level dropped. A two-volume FLASH2 model predicted a level collapse to the exit nozzle for B-19B.⁽⁵⁾ RELAP2,⁽¹¹⁾ another blowdown code, also predicted a level collapse to the exit nozzle.⁽¹²⁾ In view of these results, the mixture velocity of 4.6 ft/sec through the plate for B-19B appears near the threshold where bubble transfer begins through the plate to the upper plenum. This conclusion precludes instabilities occurring in riser and downcomer regions on the plate. This velocity represents a

FIGURE 2.15 Level of Fluid Remaining in Vessel, Run B-19B

fluid velocity against which the bubbles cease to rise and is somewhat higher than the velocities used as parameters in bubble-rise-model blowdown calculations. This velocity represents a physically real value, however, and may be useful in further refinement of multiple-volume blowdown codes.

Transient Loads on a Core Plate - M. E. Witherspoon

Strain bolts supporting the core plate are used to estimate the average differential pressure on the free area of the plate during the transient decompression. The 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. Magnitudes and waveforms obtained from these strain bolts were compared to the WATERHAMMER⁽¹³⁾ predictions for the bottom blowdown configuration in the previous quarterly report.⁽³⁾ The magnitude of the peak values predicted were found to be 3 to 5 times the observed peak values for the bottom blowdown configuration.

Additional data on strain bolt loading were obtained from the middle nozzle blowdown series. One distinct difference in the initial decompression waveform is shown in Figure 2.16. In the bottom nozzle blowdown configuration, (Figure 2.16a) the double spiked waveform is seen in the data for the first decompression wave and is less obvious in further loading cycles. This waveform is not seen in the middle nozzle data (Figure 2.16b). Further analysis is required to explain the differences in the dynamic response of the plate in the first configuration versus the second.

Data from the strain bolts for two subcooled middle-nozzle blowdowns have been consolidated in Table 2.9. The trend in this series of blowdowns is the same as that observed in the bottom blowdown series, thus indicating the equivalent pressure loading on the free area of the core plate to be less

FIGURE 2.16 Waveforms Observed in Plate Loading

than that expected for the peak differential pressure predicted using the code WATERHAMMER. Study of the mechanical response of the plate is continuing.

TABLE 2.9. *Strain Bolt Versus WATERHAMMER Comparisons*

<u>Blowdown Number and Conditions</u>	<u>Strain Gage</u>	<u>Observed Equivalent Peak Differential Pressure From Bolts, psi</u>	<u>WATERHAMMER Peak Differential Pressure, psi</u>	<u>Ratio (2/1)</u>
B-22B	SB5I	6.8	22.5	3.3
2 in.	SB6I	5.7	22.5	3.9
1300 psia	SB8I	7.7	22.5	2.9
500 °F				
		Avg. 6.7		3.4
B-25	SB5I	16.8	75.	4.5
4 in.	SB6I	18.5	75.	4.1
1300 psia	SB8I	20.2	75.	3.7
500 °F				
		Avg. 18.5		4.1

Two-Dimensional Effects During Subcooled Blowdown -
N. P. Wilburn

It has been previously noted⁽³⁾ that one-dimensional descriptions of the subcooled portions of a blowdown by codes such as WHAM do not predict all the pressure oscillations observed experimentally. In order to determine the magnitude of the multi-dimensional effects, a simplified analysis was undertaken of the two-dimensional wave equation in cylindrical geometry, and experimental equipment was designed to be installed in the CSE reactor simulator vessel to measure the pressure difference between the centerline and the periphery.⁽³⁾

During this quarter the experimental equipment was built and installed but, due to installation problems and leaks, no data have yet been obtained from the device.

An analytical solution has been obtained for the two-dimensional wave equation in cylindrical geometry for boundary conditions described in the following paragraph.

Given a right circular cylinder of length L , radius a_0 , axial dimensions Z and radial dimension r , with a circular hole of radius a centrally placed in one end ($Z = 0$), the velocity of the fluid at the hole would be a known product function of radius and time $g(r)f(t)$. This simple geometry approximates the geometry existing for the CSE vessel. The velocity function $f(t)$ is taken from a one-dimensional WATERHAMMER run for the same geometry where the hole is the opening into a pipe. The velocity function $g(r)$ is taken to be 1 for $r < a_1$ and 0 otherwise. The fitting of $f(t)$ was done by an analytical function using a non-linear least squares routine.

As a check on this approach, the corresponding analytical solution to the one-dimensional wave equation has been obtained. Using the function $f(t)$ as input, the numerical results from the solution will be compared with the WATERHAMMER run from which $f(t)$ was obtained.

Reactor Simulator Vessel Defect Surveillance -

W. C. Townsend

Ultrasonic inspections were conducted between blowdowns on the bottom hemispherical heads of the reactor simulator vessel. In addition, the small defect indication in the top head and three areas identified as possible sources of acoustic emissions were rechecked. Inspection results indicate no enlargements in the defect indication areas. No confirmation of a defect was obtained at the locations identified as sources of acoustic emission signals.

PARTICLE TRANSPORT TO SURFACES BY CONDENSING STEAM -

L. C. Schwendiman, J. M. Hales

The theoretical analysis of aerosol deposition in laminar, condensing-steam boundary layers⁽¹⁴⁾ has been completed. During the final stages, the analysis was modified to include the effects of thermophoresis. A comparison of computed particle-deposition rates, including and neglecting thermophoretic effects, showed thermophoresis to be responsible for increasing particle deposition from 1 to 9%, over the conditions studied. Results of the analysis were applied to describe the transient properties of a hypothetical vessel with a 10-ft diam and a 40-ft height. For conditions wherein the bulk atmosphere consisted of saturated steam, aerosol, air at 212 °F, and a wall temperature of 211 °F, predictions were made of

- (1) an instantaneous aerosol decay rate of 2.1%/hr,
- (2) an instantaneous steam condensation rate of 5%/hr, and
- (3) an instantaneous temperature fall of 3 °F/hr.

These predictions are expected to be lower than would be observed physically, owing to the neglect of end effects and turbulent behavior. The results of this analysis have been described in a document (BNWL-1125) to be published in the near future.

Since completion of the laminar boundary layer analysis, emphasis has shifted toward the study of particle removal through spray scrubbing. Design of an experimental system for this purpose has been initiated. Presently this system is visualized as a long vertical column wherein a stream of droplets of known size is allowed to fall through an aerosol-containing steam-air mixture. Analysis of collected droplets will allow determination of collection efficiencies. A parametric investigation is contemplated wherein the effects of varying drop size, aerosol size, bulk conditions, and droplet temperature can be ascertained.

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3. PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

CRACK DETECTION IN PRESSURE PIPING BY ACOUSTIC EMISSION

P. H. Hutton and H. N. Pedersen

Introduction

A method of continuously monitoring nuclear reactor pressure boundaries to detect formation or growth of flaws would be of significant benefit to nuclear safety. A potential method of accomplishing this is by detection of "acoustic emission" or elastic waves generated in solids by energy released during deformation and fracture. Development of the technology and equipment to apply this method to thermal power reactors is in progress at BNW. The work was started in February, 1966 under sponsorship of the USAEC's Division of Reactor Development and Technology.

Related DRDT sponsored work is in progress at Idaho Nuclear, Idaho Test Site, Idaho Falls. This phase of the work is concerned with the plant applications aspect.

Extension of the technique for application to the LMFB program is also considered in the development program. The major considerations here are the effect of higher LMFB temperatures on acoustic emission generation and on the detection system.

Task 1 - Signal Analysis and Characterization

The advantages of a sharper wave front and a more uniform propagation velocity attainable by detecting the shear wave component of acoustic emission signals have been discussed in previous quarterly reports. Use of wedge mountings to couple the shear signal from specimen surface to the sensor, discussed in the last quarterly report, is being investigated. A heavy carbon steel plate, 4 ft by 8 ft by 4 in., equipped with

bend tabs to introduce acoustic emission is being used as a test specimen in this work. Figure 3.1 shows the test arrangement.

Location of the acoustic emission source by triangulation is an important consideration in developing an acoustic emission monitor system. Triangulation accuracy should be enhanced through the use of wedge-mounted shear crystals. The wedge would be made of a material such as silver or brass with a low shear wave propagation velocity, and with the wedge angle about equal to the shear critical angle. The advantages of this method would be in the tendency of the faster traveling compressional waves and the shear waves mode converted from compressional waves to be received only with low sensitivity, thus eliminating a potential source of ambiguity in propagation velocity. Also, if the wedge angle should be optimized to receive shear waves traveling nearly parallel to the surface, then propagation velocity would be essentially constant at the shear wave velocity.

Some signal waveform comparisons have been made as a first step in evaluating a wedge-mounted shear crystal design. Rectangular PZT shear crystals were cemented to a brass wedge and the wedge cemented to the surface of a 4 in. by 4 ft by 8 ft plate at a distance of about 36 in. from one end (Figure 3.2). Alongside the wedge was a flat-mounted poled PZT crystal of 0.080-in. thick diameter with primary resonance centered at about 670 kHz. Shown in Figure 3.3 (a through d) are (a) frequency response spectrum of the wedge mounted crystal with wideband amplification, (b) same as (a) but with narrow band (600 to 800 kHz amplification), (c) flat-mounted crystal with wideband amplification, and (d) same as (c) but with narrow band amplification (600 to 800 kHz).

With amplifier gains set at approximately similar noise backgrounds, photographs were taken comparing the response of

FIGURE 3.1 *Heavy Carbon Steel Test Specimen for Acoustic Emission Study*

FIGURE 3.2 *Wedge-Mounted Shear Sensors for Acoustic Emission Detection*

(a) Wedge Mount - Wide Band

(b) Wedge Mount - Narrow Band 600-800 KHz

(c) Surface Mount - Wide Band (d) Wedge Mount - Narrow Band, 600-800 KHz
All Photos: Center Frequency = 500 KHz Frequency, Band/Division = 100 KHz

FIGURE 3.3 *Frequency Response Spectrum of Acoustic Emission Sensors to Surface Excitation*

the two sensors to excitation from an electric spark on the plate surface and from acoustic emission generated by manually bending a notched metal tab welded at the end of the plate.

Sensor responses to excitation from an electric spark are shown in Figure 3.4. The upper waveform in each photo is from the wedge-mounted sensor, and the lower waveform is from the flat-mounted sensor. Photographs (a through d) show comparisons when the wedge-mounted sensor response is limited to (a) >300 kHz and (b) >1 MHz. Photograph (c) is identical to (a) except that the spark source is located at the edge of the plate instead of the top of the plate, and (d) shows same as (a) except that the spark source is located at the underside of the plate instead of the top. In all cases, the flat-mounted sensor response was limited to >700 kHz. These photos show both crystals to be somewhat sensitive to energy arriving at velocities faster than pure shear velocity. Both crystals are more sensitive to the slower traveling shear energy and, to a lesser degree, other late arrivals, i.e. those combinations of mode conversions and multiple reflections. The wedge mounted crystal in this experiment showed more sensitivity at the edge and bottom source locations than did the flat mount sensor. However, it also had an advantage of being tuned to accept lower frequency components.

Some comparisons of response to acoustic emission generated by bending tabs at the end of the plate are shown in Figure 3.5. Figure 3.5a compares the wedge-mounted sensor to the flat-mounted sensor when the signal from the wedge-mounted sensor is amplified at a narrow band frequency of $\Delta f_w \approx 230 - 340$ kHz, whereas the signal from the flat-mounted sensor is amplified at a narrow band frequency of $\Delta f_f \approx 600$ to 800 kHz. Points of comparison in Figure 3.5 (b through f) are as follows:

(b) same as (a) except $\Delta f_w \approx 340 - 510$ kHz;

(a) Top: Wedge Mount > 300 KHz
Bottom: Surface Mount > 300 KHz
Excitation: Top of Plate at End

(b) Top: Wedge Mount > 1 MHz
Bottom: Surface Mount > 700 KHz
Excitation: Top of Plate at End

(c) Top: Wedge Mount > 300 KHz
Bottom: Surface Mount > 300 KHz
Excitation: Edge of Plate at End

(d) Top: Wedge Mount > 300 KHz
Bottom: Surface Mount > 700 KHz
Excitation: Underside of Plate at End

Sweep Rate For All Photos: 50 μ sec/cm

FIGURE 3.4 Comparisons of Wedge Mounted Shear Sensor and Flat-Mounted Sensor Responses to Electric Spark Excitation

(a) Top: Wedge Mount, 230-340 KHz
Bottom: Surface Mount,
600-800 KHz

(b) Top: Wedge Mount, 340-510 KHz
Bottom: Surface Mount,
600-800 KHz

(c) Top: Wedge Mount, 510-760 KHz
Bottom: Surface Mount,
600-800 KHz

(d) Top: Wedge Mount, 760-1140 KHz
Bottom: Surface Mount,
600-800 KHz

(e) Top: Wedge Mount, 1140-1700 KHz
Bottom: Surface Mount,
600-800 KHz

(f) Top: Wedge Mount, 1700-2560 KHz
Bottom: Surface Mount,
600-800 KHz

Sweep Rate For All Photos: 50 μ sec/cm

FIGURE 3.5 Comparisons of Wedge-Mounted Shear Sensor and Flat-Mounted Sensor Responses to Acoustic Emission

- (c) same as (a) except $A_{fw} \approx 510 - 760$ kHz;
- (d) same as (a) except $A_{fw} \approx 760 - 1140$ kHz;
- (e) same as (a) except $A_{fw} \approx 1140 - 1700$ kHz; and
- (f) same as (a) except $A_{fw} \approx 1700 - 2560$ kHz.

These photographs demonstrate good sensitivity for the wedge-mounted sensor up to about 1.5 MHz. At higher frequencies, the transducer becomes more vulnerable to compressional wave components and less sensitive overall. Due to changing tabs positions but not to change in the scope trigger sensor location, the time position of the received signals in the photographs do not bear a constant relationship to propagation velocity.

The wedge-mounted crystal shows some promise for applications service, but different crystals must be obtained to improve the noise-free high frequency response range.

The wedge technique offers a method of obtaining the advantages of high frequency shear sensors for acoustic emission detection on a practical basis, i.e., it does not require access to the edge of the material. Further work is required, however, to improve wave front definition.

Definition of the maximum range at which acoustic emission signals can be reliably detected in various frequency ranges is essential to defining the complexity of a full scale monitor system for on-line surveillance. Detection frequencies appropriate for various conditions and the attendant equipment are sufficiently resolved to make a study of detection range timely at this point. Work is in progress to define detection range in terms of at least two parameters consisting of sensing frequency and material thickness. Work to date has been done using the 4-in. thick plate shown in Figure 3.1. Types of sensors studied so far are thickness-poled, air-backed, encased PZT-5A crystals of 0.080, 0.120, and 0.240-in. diameters, and lithium niobate shear crystals. The

thickness-poled sensors were mounted flat on the plate surface and the shear sensors were mounted on the edge of the plate using an epoxy couplant. Figure 3.6 shows installed sensors.

Study work to date indicates response attenuation of the surface-mounted PZT-5 sensors to be about 3 db/ft. Reliable detection range was limited from 4 to 5 ft. The larger acoustic emission signals were detectable at greater distances, but the proportion of the total number of signals detectable at the greater distances was less than 50%. Response to acoustic emission of various surface-mounted sensor samples is shown in Figure 3.7 to be at distances of 3 and 4 ft from the source.

The lithium niobate shear sensor showed good response to acoustic emission at 8 ft from the source. Since this is the maximum dimension of the plate, no response for distances greater than 8 ft could be measured in this test. A comparison of shear sensor response at 8 ft to a surface-mounted sensor response at 3 ft to the same emission signal is given in Figure 3.8.

Duration of the original signal appears to effect detection distance particularly with the surface-mounted sensors. A very short signal does not propagate as far as a longer duration signal with the same maximum amplitude. This observation may relate to the generation of standing waves in the specimen. Further study of detection range is needed to arrive at a final conclusion.

Task 2 - Sensor Development

A practical method of calibrating acoustic emission sensors in terms both meaningful and directly comparable among the various sites performing acoustic emission work has been a problem. A technique for calibrating the frequency response and relative sensitivity of acoustic emission sensors has been developed using an air abrasive unit to generate broad spectrum noise in a test block or specimen to which the

**Surface Mounted Sensors
Thickness-Poled**

Edge-Mounted Shear Sensors

FIGURE 3.6. *Sensors Used to Study Acoustic Emission
Detection Range*

0.080 in. Diam, 500 KHz Hi-Pass

0.120 in. Diam, 500 KHz Hi-Pass

0.240 in. Diam, 300 KHz Hi-Pass

All Photos: 100 μ sec/cm horizontal, 0.05 V/cm vertical

FIGURE 3.7 *Surface Mount, PZT-5 Sensor Response to Acoustic Emission (Sensor Size and Distance to Source Varied)*

transducer is bonded. The frequency response curve and relative sensitivity of the transducers are measured with a spectrum analyzer or tunable voltmeter.

Lithium Niobate Shear
8 ft from Emission
source, 0.02 V/cm.

0.080-in. Diam PZT-5
Sensor 3 ft from
Emission Source 0.05 V/cm.

100 μ sec/cm 500 KHz Hi-Pass

FIGURE 3.8. *Comparative Response of Lithium Niobate Shear Sensor and 0.080 in. Diam PZT-5 Surface Mount Sensor to the Same Acoustic Emission Signal*

The calibration technique has been developed around an S. S. White Model F air abrasive unit used in the transducer laboratory for cutting and shaping piezoelectric transducer elements. This unit uses a magnetic vibrator to control the abrasive flow rate. The flow is adjusted by varying the voltage on the vibrator. The abrasive is carried through the nozzle by a high velocity air stream. A number of abrasive particle sizes are available. However, spectral responses obtained to date have been taken with 50 μ alumina powder.

The response curves are measured on a Hewlett Packard 8552A/8553L Spectrum Analyzer. A 10 kHz bandwidth is used, and the spectrum analyzer output is integrated for display

of an average reading on a scope. This average reading is corrected to obtain a true rms value. Initially, the peak output, displayed directly on the scope, gave the appearance of a "grassy" response curve. For any given transducer, the peak output response has the same shape as the averaged response.

A number of response curves for a single transducer were taken to study the effect of operating parameters such as air pressure, abrasive flow rate, etc. on the response curves. This transducer has a 0.070-in. diam, PZT-5A, 2-MHz element poled in the longitudinal mode. The element is bonded into the case with Budd GA-100 strain gage adhesive, and the case is filled with Sauereisen cement.

Several factors were investigated for their influence on the acoustic spectrum generated by an abrasive stream. Pressure and abrasive flow rate appear to affect the amplitude but not the spectrum shape. The material in the test block or specimen and the abrasive particle size appear to affect the shape of the acoustic emission spectrum to a limited degree. The relative positions of the transducer and the abrasive stream, as well as specimen geometry appear to have no effect.

Figure 3.9 shows the characteristic response pattern obtained for four different sized acoustic emission sensors using the air abrasive unit as an excitation source. This technique offers a simple, reproducible method of relating the response characteristics of all types of acoustic emission sensors.

As discussed in previous quarterly reports, lithium niobate piezoelectric material offers the most promise for obtaining an uncooled acoustic emission sensor with temperature capability above ~ 625 °F. Previous tests of the various

0 1 2 0
Frequency, MHz
0.070 in. diam, **PZT-5**
Epoxy Backed

0 1
Frequency, **MHz**
0.080 in. diam, **PZT-5**
Air Backed

0 1 2 0
Frequency, MHz
0.120 in. diam, **PZT-5**
Air Backed

0 1 2
Frequency, MHz
0.240 in. diam, **PZT-5**
Air Backed

FIGURE 3. 9. *Typical Calibration Spectra for Acoustic Emission Sensors with Air Abrasive Unit as Excitation Source*

sensor configurations using this material in the compressional mode have shown very marginal sensitivity for acoustic emission detection. Lithium niobate sensors operated in the shear mode have been included in emission detection range tests earlier in this report. It is very encouraging to note that the sensitivity in this mode appears adequate to detect acoustic emission. Figure 3.8 illustrates the comparative response of a lithium niobate shear sensor at 8 ft from an acoustic emission source, versus a thickness-poled PZT-5 surface-mounted sensor at 3 ft.

The first in a series of tests to evaluate the effect of nuclear radiation on acoustic emission sensor crystals has been completed. This test involved exposing PZT-5A crystals to a ^{60}Co gamma source (1.33 and 1.17 MeV) at room temperature. No change could be detected in the receiving or transmitting characteristics of the crystal up to an accumulated exposure of $4.9 \times 10^8 \text{R}$ at the point of test termination. This exposure was well above the estimated minimum requirements for acoustic emission sensors used in a continuous, on-line monitor system for thermal reactors.

Task 3 - Signal Conditioning System

Preamplifiers used in an acoustic emission monitor system may be exposed to temperatures in the vicinity of 200 °F because of the need to locate them near the sensors. The standard tuned preamplifier has been tested to evaluate its resistance to temperature damage. Results show that the standard unit is capable of operating for extended periods at 185 °F. The results also indicated that the service temperature could be increased to 200 °F by simply substituting high temperature wiring for the standard wiring now used.

Interference from intense low frequency noise is a concern in attempting to detect acoustic emission on an operating

system such as a reactor pressure system. A high pass filter with 36 db/octave attenuation below the pass point, and with selectable pass points of 300, 500, 800 kHz and 1 MHz, has been developed and tested. Since the filters previously used were limited to 24 db/octave, this represents a significant improvement in noise suppression.

Task 4 - Data Analyzer and Readout System

No significant developments.

Task 5 - Application Engineering

Acoustic emission detection has been applied locally to investigate potential slow crack growth by fatigue in areas of known high stress in reactor process piping. Of the five locations monitored, three showed indications of limited crack activity. Subsequent ultrasonic examination, however, failed to confirm the existence of a crack. The decision has been made on the basis of total engineering information to replace all of these sections of pipe. Arrangements to salvage and to examine in detail those sections producing emission indications have been made for the purpose of establishing the source of the acoustic emission. This approach potentially may contribute significantly to establishing a criterion for evaluating acoustic emission indications.

Surveillance of known flaws in the heads of the Containment System Experiment simulator vessel by acoustic emission detection has continued. Trial modification of the monitor system to operate at 600 kHz and above instead of the original 200 kHz and above showed significant improvement in elimination of flow noise from the data. Acoustic emission indications continue to be very limited.

Task 6 - Cooperative Work with Idaho Nuclear

Verbal liaison only.

General

A paper titled "Detecting Acoustic Emission in the Presence of Nuclear Reactor Hydraulic Noise" was presented at the 15th annual meeting of the American Nuclear Society at Seattle, Washington, June 15-19, 1969.

Work Planned for Next Quarter

- Continue work on adaptation of the shear sensor to continuous surfaces.
- Continue study of maximum acoustic emission detection range.
- Continue development and testing of the lithium niobate shear sensor, including irradiation testing.
- Investigate high temperature plastic molding process for sensor fabrication.
- Investigate the effect of temperature on attenuation of acoustic emission signals.
- Investigate the effect of temperature on generation of detectable acoustic emission in both stainless and carbon steel.

4. ENVIRONMENTAL STUDIES

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

Operations research with the COL HEAT thermal simulation model to investigate the potential effects of an expanding power industry is underway. The purpose of the study is to simulate the thermal regimes of major streams of the United States and to subsequently simulate the average and minimum capacities of the streams, within a given set of constraints, for direct cooling of generating plant condensers. The constraints will include the temperature restrictions imposed by State Water Quality Standards and the economic limitations on minimum plant capacity, as well as the restrictions imposed by the nature of the stream, weather, and solar radiation. Initially, two previous test studies, the Ohio Basin and the Upper Mississippi Basin, are being reevaluated to conform to the indicated format. These studies were originally carried out to ascertain whether the COL HEAT model, first developed and tested on the Columbia River, could be utilized to simulate the thermal regimes of other major river basins.

During the report period the following accomplishments were made:

- A satisfactory simulation of the hourly thermal regimen of the Phelps County (Nebraska) Canal was achieved by desensitizing regional weather variables. Weather and **radiation** data from stations located at distances of approximately 100 miles were utilized as rolling means. As a result, a series of point measurements made on site in 1968, including the thermal input of the 100 MW Canaday Generating Plant operated by the Central Nebraska Power and Irrigation Cooperative, was fitted to within 1 °C of computed values. In order to ascertain the effect of increased thermal loads on the

canal, the observed load was increased by factors of 3 and 5. The results of this simulation are illustrated in Figure 4.1. A report on the full study is in preparation.

Simulation of the 1964 thermal regimen of the Ohio River Basin, including the Ohio River Main Stem and major tributaries, was completed. Early difficulty in simulating upstream sectors was resolved when it was determined that Huntington, West Virginia weather was more representative of local conditions at most upstream points than was Pittsburgh weather. The dominant influence of westerly winds appeared to be the important factor.

- Simulation of the thermal capacity of Ohio Basin streams was carried out under the constraints of average monthly flow in the 1950-60 decade, average long term monthly weather and radiation, a maximum river temperature increase of 5 °F, and a minimum thermal discharge of 800 MW. A 5 °F increase is a common but not universal limitation established by State Water Quality Standards; while the 800 MW minimum corresponds approximately to the effect of a 500 MW nuclear unit operating at a capacity factor of 80%. The results of these studies on two tributary streams, the Allegheny River near Pittsburgh and the Muskingum River of Ohio, are illustrated in Figures 4.2 and 4.3. Figure 4.3 indicates, for example, that a 500 MW nuclear generating unit could be expected to cause an increase of greater than 5 °F, in the lowermost 93 miles of

Distance Below Canady Outfall, miles

FIGURE 4.1 Phelps County, (Nebraska) Canal

FIGURE 4.2 Allegheny River, from River Mile 50 to River Mile 0

FIGURE 4.3 Muskingum River, from River Mile 93 to River Mile

the Muskingum River. This expected increase would be for an average of 3 months in the late summer and early fall. Similar information has been developed for the entire Ohio Basin. These data have been analyzed in terms of projected power growths and will be included in the forthcoming Ohio Basin Report. The effects of extreme conditions of weather and river flow were not included in this study of the Ohio Basin. They are to be included, however, in the continuing study now underway.

- A thermograph at Alton, Illinois (Lock & Dam 26) on the Upper Mississippi River was replaced after being damaged during the previous report period. Two additional thermographs were placed in operation at Fulton, Illinois (L&D 14) and at Welch, Minnesota (L&D 3) to provide improved data for simulation of the Upper Mississippi River. Three units are also planned for installation on the lower river.

5. FIXATION OF RADIOACTIVE RESIDUES

OVERALL STATUS OF WSEP DEMONSTRATIONS - K. J. Schneider

Table 5.1 shows the overall status of the radioactive demonstrations in WSEP as of July 31, 1969. A total of 23 demonstrations have been completed, 6 in the pot calcination process, 7 in the phosphate glass solidification process, and 10 with the spray solidification process. Approximately 32 MCi of mixed radionuclides representing waste from about 240,000 MWd of electrical power have been converted to solids. This amount of waste represents 91 days of all the nuclear power capacity in the United States as of March 1, 1969. The total on-line processing time for solidification of these wastes was 58 days.

TABLE 5.1. *Overall Status WSEP Radioactive Demonstrations*

	Solidification Method			Total
	Pot	Spray	Phosphate Glass	
Runs Completed	6	10	7	23
Megacuries Solidified	4.0	17.5	10.6	32
Equivalent Tonnes ^(a) Processed	11.3	14.6	10.1	36
MWd _e Represented by Waste ^(b)	75,000	98,000	67,000	40,000
Tonne/Day Rate ^(a)	0.6 to 1.0	0.5 to 0.9	0.3 to 0.7	--
Maximum kW in One Pot	5.1	12.7	9.8 ^(c)	118 ^(e)
Maximum W/ℓ in Pot	85	205	160 ^(d)	--
Maximum Centerline Temperature in Pot, °C	940	930	780	--
Liters Solid/Tonne ^(a)	40 to 50	30 to 65	50 to 100	--

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

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

(c) True maximum has not yet been demonstrated. Calculated maximum is 11.7 kW.

(d) True maximum has not yet been demonstrated. Calculated maximum is 195 W/ℓ.

(e) Total kilowatts encapsulated to date.

SPRAY SOLIDIFICATIONENGINEERING STUDIESDevelopment Spray Solidification Runs - J. D. Moore

During nonradioactive development and WSEP spray solidification runs with PW-4m feed, between 20 and 60% of the ruthenium evolves from the feed during calcination. To determine if the ruthenium evolution from the solidifier could be reduced, four runs were made with the development spray solidifier to test methods of ruthenium retention. Run data are summarized in Table 5.2.

TABLE 5.2. *Summary of Development Spray Solidification Runs*

	<u>DSC-31</u>	<u>DSC-32</u>	<u>DSC-33</u>	<u>DSC-34</u>
Feed Type	PW-4m	PW-4m	PW-4m	PW-4m
Feed concentration, liters/tonne	506	454	454	378
Additives	Fe,Al, Na,Li, H ₃ PO ₄	ZnO, Ca(OH) ₂	ZnO, Ca(OH) ₂	HNO ₃
Feed total acidity, M H ⁺	2.4	Basic pH=10	Basic pH=10	1.7,3.4, 5.1
M ⁺ /P	2.5	--	--	--
Feed consumed, liters	125	77.2	31.9	303.9
Average feed rate, liters/hr	15.6	18.4	16.8	15.5
Calciner furnace temperature, °C	700	700	700	700
Calciner vacuum, inches of water	8	5	5	8
Atomizing gas	air	air	air	air
Atomizing gas flow, scfm	7	6.9	6.5	6.5
Filter blowback pressure, psig	40	40	40	30
Filter pressure drop, end of run, inches of water	10	5	7	4
Melter furnace temperature, °C	800	--	--	--
Product collected, kg	27.3	17.3	3.6	48
Product concentration, liters/tonne	32	--	--	--
Particulate DF (feed to condensate)	660	500	NA	NA
Ruthenium volatilized, % of total feed ruthenium	38	<2	NA	<2, <0.6, <3

NA - Not Available

Prior to Run DSC-31, PW-4m feed (the same flowsheet used in WSEP Runs SS-9 and 10) containing phosphate flux (see Table 5.3 for detailed feed compositions) was acid-stripped from a total acidity of 6.05M to 2.4M. Previous experience with PW-1 feeds had shown that ruthenium release increased with feed acidity." One hundred and sixty-eight liters of feed were cycled through a horizontal wiped-film evaporator at an average rate of 95 liters/hr during an intermittent 53 hr run in preparation for spray solidification. When the feed was processed in DSC-31, 38% of the ruthenium was volatilized to the spray solidifier off-gas system. The above reduction in feed acidity did not significantly reduce the ruthenium evolution, and apparently a much more extensive acid strip would have been required to do so.

Since acid-stripping is very time consuming with the available evaporators, both in the nonradioactive laboratory and in the WSEP, chemical neutralization of the feed was tried. Simulated PW-4m was made basic (pH of 10) by adding Ca(OH)_2 (0.65M) and ZnO (0.5M). When diluted to 454 liters/tonne, the feed contained about 70 vol% solids (the precipitate was a fine, easily suspended solid), but a relatively steady feed flow was nevertheless achieved in Runs DSC-32 and 33. No flux was added to the feed, so the calcine was collected in a barrel connected to the bottom of the calciner. Less than 2% of the ruthenium was volatilized from the feed during these runs.

In DSC-34 the effect of feed acidity on ruthenium retention in the absence of phosphate ion was studied. Three PW-4m feed batches were prepared with total acidities of 1.7M, 3.4M, and 5.1M, respectively. No additives other than

*BNWL-1003, 68-4.

TABLE 5.3. *PW-4m Feed Composition for Development Spray Solidifier Runs*

<u>Chemicals</u>					
Fe ⁺³					0.124M ^(a)
Cr ⁺³					0.012
Ni ⁺²					0.051
Al ⁺³					0.001
Na ⁺					0.342 ^(b)
SiO ₃ ⁻²					0.01
PO ₄ ⁻³					0.003
NO ₃ ⁻					1.932
H ⁺					0.037
<u>Fission Products</u> ^(e)					
MoO ₄ ⁻²					0.161
Sr ⁺²					0.036
Ba ⁺²					0.041
Rb + Cs (used K ⁺)					0.092
(Y + RE) ⁺³					0.274
ZrO ⁺²					0.143
Ru ⁺⁴					0.0082 ^(c)
Rh (used Co ⁺²)					0.013
Ag ⁺					0.0016
Cd ⁺²					0.0025
Te (used SO ₄ ⁻²)					0.014
Pd (used Ni ⁺² , included above)					0.043
Te (used MoO ₄ ⁻² , included above)					0.031
<u>DSC-34</u>					
<u>Additives</u>	<u>DSC-31</u>	<u>DSC-32-33</u>	<u>Part 1</u>	<u>Part 2</u>	<u>Part 3</u>
Fe ⁺³	0.9	--	--	--	--
Al ⁺³	0.2				--
Li ⁺	0.75	--	--	--	--
Na ⁺	0.75				
PO ₄ ⁻³	2.8			--	--
Ca ⁺²	--	0.65			
ZnO		0.5	--	--	--
OH ⁻	1.5	1.3	--	--	--
H ⁺	5.6 ^(d)		0.83	2.33	3.83
NO ₃ ⁻	3.3 ^(d)	--	0.83	2.33	3.83

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

(b) Nominal Na concentration is 0.1M. Excess is from using Na₂MoO₄·2H₂O for Mo addition.

(c) Nominal Ru concentration is 0.082M.

(d) Concentrations before acid-stripping.

(e) Simulated with nonradioactive chemicals.

the nitric acid necessary to adjust the feed acidities were used. About 100 liters from each batch were fed to the spray calciner. Powder was collected in a product pot connected to the calciner. Less than 3% of the ruthenium was evolved from all three feed batches with no significant difference in the amount released from any of the three.

From the results of these tests, it appears that ruthenium can best be retained in the product if the phosphate flux is added as a separate stream to the melter. Since the ruthenium evolution from phosphate-free liquid feeds is minimal with acidities as high as 5.1M, acid-stripping of the radioactive waste during feed preparation would not be necessary. Further flowsheet development is necessary, however, to find the type and amount of flux necessary to form a good solid product.

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

Spray Solidification Run SS-10

During Run SS-10, a simulated waste equivalent to that resulting from the reprocessing of 1.23 tonne of power reactor fuel (45,000 MWd/tonne at 30 MW/tonne) with an equivalent aging time of 1.8 years was reduced to 58 liters of ceramic product. The radioactive ceramic produced a heat rate density of 127 W/liter (7370 W total based on pot calorimetry) in an 8-in. diameter mild steel melt receiver. The steady-state pot centerline temperature in the air-cooled furnace was 611 °C, and the centerline-to-wall temperature difference was 262 °C. These values indicate an effective thermal conductivity of $1.25 \text{ W/(m}^2)(^{\circ}\text{C/m)}$ [$0.72 \text{ Btu/(hr) (ft}^2)(^{\circ}\text{F/ft)}$] for the ceramic product.

The solidifier was fed 750 liters of PW-4m feed [additives of 0.9411L $\text{Fe}(\text{NO}_3)_3$, 0.16M $\text{Al}(\text{NO}_3)_3$, 0.75M LiOH , 0.97M NaOH , and 2.80M H_3PO_4 based on 378 liters/tonne] at 611 liters/tonne in 56 hr at an average feed rate of 13.5 liters/hr. The feed contained 2,580,000 Ci of radioactivity including 12,700 Ci of ruthenium. A summary of run data for SS-10 is presented in Table 5.4.

TABLE 5.4. *Summary Data for WSEP Spray Solidifier Run SS-10*

<u>Operational Data</u>	
WSEP Run	22
Date	4/69
Operating Mode	67% A - 33% B
Feed Type	PW-4m
Feed, liters/tonne	611
Pot Diameter, in.	8
Pot Material	Mild Steel
Total Run Feed Time, hr	55.5
Average Feed Rate, liters/hr	13.5
Total Feed, liters	750
Total Feed, tonne equivalent	1.23
Feed: Solid Volume Ratio(a)	11.4
Feed: Solid Volume Ratio at(a) 378 liters/tonne Basis	7.1
Filter AP, in. of water	5-9

Problems

Deposition of calcine in the bottom conical section of calciner and filter chamber.

<u>Filled Pot Data</u>	
Total Curies in Feed to Pot ^(b)	2,270,000

TABLE 5.4. (Contd)

Total Curies of Ru in Feed to Pot(b)(c)	6,150
Solid in Pot, liters	58
Solid in Pot, liters/tonne(a)	55
Heat in Pot, W(d)	7,370
Equivalent Aging Time, years	1.8
£ Temp in Air-cooled Furnace, °C	611
£ to Wall A Temp., °C	262
Effective Thermal Conductivity, W/(m ²)(°C/m)[Btu/(hr)(ft ²)(°F/ft)]	1.25[0.72]
Welded Lid	Yes
Radiation Reading, R/hr at 6 in.	25,000

(a) *Twelve percent of product assumed retained in solidifier.*

(b) *By feed analysis and assuming 12% of activity retained in solidifier.*

(c) *Forty-five percent of total ruthenium volatilized from solidifier.*

(d) *By pot calorimetry data.*

During the run, the flow of calcine from the spray calciner into the melter was difficult to maintain. The flow was more erratic than in previous runs, and occasional chunks of calcine were noted entering the melter, particularly during filter blowback. Early in the run the feed was interrupted for 3-1/2 hr to rod out what was thought to be a partial calcine bridge in the conical bottom outlet of the calciner. However, the rodding dislodged very little calcine, and increased vibration of the calciner was required for the remainder of the run to maintain calcine flow to the melter. The filter pressure drop also increased from 5 to 9 in. of water during the run. Raising the filter blowback pressure from 40 to 60 psig did not stop the increasing trend in

filter pressure drop, but it may have slowed it. This is the first time during the last seven spray solidification runs that a significant increase in filter pressure drop has occurred during a run. An inspection of the inside of the solidifier was made to determine, if possible, the cause of the difficulties experienced with the calcine flow and filter pressure drop. Two calcine deposits were found in the conical bottom section of the calciner; the main deposit was located in the area directly below the calciner barrel and a smaller one was located in the area below the filters. Considerable deposition was noted in the filter chamber as caked calcine was lodged around the bottom section of the filters. This explains the erratic calcine flow during filter blowback and the increase in filter pressure drop during the run. The calciner walls were relatively clean. In all cases, the deposits appeared to be the result of wet calcine depositing and drying to form a cake. This suggests inadequate feed atomization, the reason for which is currently under investigation. Based on calorimetry tests of the pot (7370 W by calorimetry versus 8350 W by feed analysis) and volume reduction data from Run SS-9*, it is estimated that approximately 12% of the feed or about 20 kg of calcine accumulated in the calciner during the run. A thorough flush of the system will be required prior to the next spray solidifier run.

The PW-4m melt was batch discharged from the melter via the freeze-drain valve into the unheated mild steel receiver. The melt was quite viscous as in Run SS-9*, but an adequate fill of the receiver was achieved.

Prior to the run, the simulated PW-4m waste, including additives, was acid stripped for approximately 50 hr in

*BNWL-1074, 69-1

the waste evaporator (TK-113) to determine the effect of reduced acidity on ruthenium volatility from the spray solidifier. As a result of the stripping operation, the total acidity of the feed was reduced from 7.5M to 3.2M (based on 378 liters/tonne). A greater reduction of the total acidity had been planned, but due to frequent plugging of the evaporator dip tubes the stripping operation was curtailed prematurely.

During the last third of Run SS-10, operation was changed from Mode A to Mode B by feeding Purex high level waste (1WW) from the evaporator feed tank (TK-112) to the waste evaporator (TK-113). Figure 5.1 shows the Mode A and B arrangement for SS-10. The activity distribution during each mode of operation and for the total run is presented in Table 5.5. Forty-five percent of the ruthenium in the feed processed during Mode A was volatilized from the solidifier and indicates that the reduction of the total acidity of the feed prior to the run did not significantly reduce ruthenium volatility from the solidifier. Only 0.08% of the CePr-144 representing the nonvolatiles in the feed processed during Mode A was entrained from the solidifier. Although Table 5.5 indicates some differences in distribution of activity downstream of the evaporator (TK-113) during Mode B, these differences are not considered significant based on data from previous Mode A runs.*

During SS-10 work continued on gas sampling techniques that will permit better definition of the type, form, and extent of activity present in the process off-gas stream. The gaseous effluent from the scrubber (TK-118) was sampled for both particulate and volatile forms of activity by routing a side stream of the effluent through first glass fiber filters

*BNWL-1074, 69-1 and BNWL-1003, 68-4.

FIGURE 5.1. Mode A and B Arrangement for SS-10

TABLE 5.5. Activity Distribution for Run SS-10

Fraction of Feed Radioactivity to Solidifier During each Mode of Operation (d) and Total Run in the Effluent Streams						
	Mode A		Mode B		Total Run	
	(Ru)	(T-Ru)	(Ru)	(T-Ru)	(Ru)	(T-Ru)
Evaporator Bottoms (TK-113) (a)	4.5×10^{-1}	7.9×10^{-4}	2.1	2.4×10^{-1}	1.0	7.9×10^{-2}
Fractionator Bottoms (TK-115)	4.0×10^{-3}	2.3×10^{-7}	7.8×10^{-3}	7.1×10^{-7}	5.3×10^{-3}	3.9×10^{-7}
Fractionator Con- densate (TK-116) (b)	0.6×10^{-6}	ND	13.0×10^{-6}	5.6×10^{-10}	4.8×10^{-6}	1.9×10^{-10}
Off-Gas to Scrubber	3.4×10^{-5}	2.1×10^{-10}	2.7×10^{-5}	$< 17.0 \times 10^{-10}$	3.2×10^{-5}	$< 7.0 \times 10^{-10}$
Off-Gas to Stack	---	---	---	---	$< 2.0 \times 10^{-10}$	1.5×10^{-12}
Ratio Fractionator Condensate Activity to 10CFR20	---	---	---	---	4.9×10^3	1.3×10^2
Ratio Stack Activity to 10CFR20	---	---	---	---	$< 4 \times 10^{-4}$	3.4×10^{-3} (c)

(T-Ru) Fraction of total feed activity less ruthenium

ND Not Detected

(a) Fraction of feed activity for Mode B and Total Run includes the activity fed to TK-113 from from TK-112 during Mode B.

(b) Essentially all the condensate was accumulated during the Mode B portion of the run and accounts to a large extent for the higher fraction accumulated during Mode B.

(c) For gross beta conservatively calculated as Sr-90 and Y-90.

(d) See Figure 1 for Mode A and B arrangement.

and then a 2M KOH solution. Essentially all of the activity was deposited on the filters. Calculations suggest that an order of magnitude for the Ru-106 concentration in the process off-gas was 10^{-8} $\mu\text{Ci}/\text{cm}^3$, the same as that observed during SS-9. The CePr-144 concentration was calculated to be about 10^{-7} $\mu\text{Ci}/\text{cm}^3$, about 100 times higher than that observed during SS-9.

During SS-10 the process off-gas stream was also sampled downstream of the process condenser (E-111) using a technique similar to the one discussed above, i.e., a portion of the off-gas from the condenser was bypassed first through a glass fiber filter and then bubbled, via a fitted plastic cylinder, through a 2M KOH solution. The Ru-106 activity associated with five different filter-caustic scrubber sample package assemblies is listed in Table 5.6. Calculations, based on the total Ru-106 activity deposited in those five sample packages, account for over 100% of the ruthenium activity that accumulated in the fractionator (about 70 Ci). Sampling of the gaseous effluent streams will continue during the phosphate glass demonstration runs.

TABLE 5.6. *Ru-106 Activity Found in E-111 Off-Gas
Sample Packages During SS-10*

Sample Package No.	Sampling Time, hr	Ru-106 Deposition Rate, $\mu\text{Ci}/\text{hr} \times 10^{-4}$	
		On Filter	In Caustic
2	2.0	2.05	0.06
3	0.8	2.40	ND
4	2.0	0.07	0.18
5	1.0	0.07	0.90
6	1.0	0.50	0.30

ND *Not Detected.*

PHOSPHATE GLASS SOLIDIFICATIONLABORATORY STUDIESCorrosion Studies - R. F. Maness

A laboratory-scale phosphate glass run was made using a PW-2 feed solution containing 0.5M NH_4NO_3 . The run was made at 1000 °C with semicontinuous feed addition to an alumina crucible equipped with an overflow weir. Total time at temperature was 33 hr, whereas feed was added semicontinuously for 22 hr. Platinum specimens were exposed to the melt phase, to the melt-vapor interface, and to the vapor phase. The platinum specimens exposed to the melt phase and to the interface were unattacked. The end of the specimen exposed to the vapor phase (about 6 in above the melt) was covered with melt as a result of splattering. Removal of the melt revealed a crack, presumably as a consequence of embrittlement under reducing conditions. No attack or embrittlement of other areas of the coupon was noted.

Platinum specimens were exposed to molten (decomposing) $(\text{NH}_4)_2\text{HPO}_4$ at 400, 600, and 800 °C and to $\text{NH}_4\text{H}_2\text{PO}_4$ at 450 and 700 °C for 16 hr periods. Corrosion was slight and no embrittlement was noted.

RADIOACTIVE DEMONSTRATIONPhosphate Glass Run PG-7 - J. L. McElroy, R. F. Drager*

During Run PG-7 simulated waste equivalent to 1.1 tonnes of power reactor fuel (20,000 MWd/tonne at 15 MW/tonne) with an equivalent aging time of 0.5 years was solidified

*On assignment from Brookhaven National Laboratory.

to produce 182 kg of glass product. The 68 liters of radioactive glass produced a heat rate density of 144 W/liter (9800 W total based on a pot calorimetry test). The heat rate density was only 80% of the planned thermal maximum that would have produced a molten core of one-half the diameter in the 8-in. diameter receiver pot.

Because of the low remelt temperature of the phosphate glass (600 to 700 °C), and because of an increased potential for corrosion, this molten core limit was established in addition to the 900 °C centerline limit and a 427 °C wall temperature limit. The steadystate pot centerline temperature of 749 °C with the pot hanging in 40 °C air indicated that the pot centerline was slightly molten. In air, a pot centerline-to-wall temperature difference of 418 °C gave an effective thermal conductivity of $0.90 \text{ W/(m}^2)(^{\circ}\text{C/m)}$ [$0.52 \text{ Btu/(hr)(ft}^2)(^{\circ}\text{F/ft)}$]. During processing, with the pot in the heated receiver furnace, the pot centerline temperature was 853 °C, and tile centerline-to-wall temperature difference was 202 °C. For this condition of the nearly completely molten glass, the effective thermal conductivity increased to $1.85 \text{ W/(m}^2)(^{\circ}\text{C/m)}$ [$1.07 \text{ Btu/(hr)(ft}^2)(^{\circ}\text{F/ft)}$].

The denitrator was fed 793 liters of adjusted sulfate containing PW-2 type feed (644 liters/tonne) in 44 hr at an average feed rate of 18 liters/hr. Approximately 100 liters of the feed remained in the denitrator at the end of the run. The feed contained approximately 2,700,000 Ci of radioactivity. A commercial silicone antifoam agent was added continuously to the denitrator as in previous phosphate glass runs*, but was not completely effective. Foaming that occurred in the denitrator was initially due to a sour batch

*BNWL-834, 68-1.

of antifoam, but foaming was still present when a fresh batch of antifoam was used. No serious operating problems resulted from the foaming.

Some plugging difficulties in the airlift pot recirculation line to the denitrator caused periods of excessive feed rates to the melter and resulted in foaming of the melt within the melter. On one occasion the foaming led to a slight pressurization of the melter when foam levels reached the melter viewing window and entered the off-gas line and the overflow seal pot line. The melter control temperature that was used to manually control the airlift feed rate to the melter did not always reflect the true internal melter operating conditions. The thermocouple is located near the melt surface in the melter. It would correctly indicate a period of overfeeding by a decrease in temperature and it would normally indicate a period of recovery by an increase in temperature. However, at times, increases in temperature were solely due to rising levels of foam which resulted from overfeeding. False assumptions of melter recovery during these periods then led to a continuation of resumption of feed to the melter and aggravated the foaming condition.

With the melter operating at a furnace temperature of 1250 °C, the average melt pool temperature was nearly 1150 °C, and the melt rate to the receiver pot was 1.45 liters/hr. The melt was overflowed through the unheated melter weir until the end of the run when it was batch-dumped through the freeze valve into the receiver pot. This was done to completely empty the melter. Receiver temperatures were controlled at approximately 600 °C to prevent possible stalagmite formation.

Initial startup of Run PG-7 was delayed by failure of the melter furnace during heatup. Although the exact cause of failure has not been found it was determined that two of the three Kanthal A-1 heating elements were open. Visual inspection of the elements revealed an extreme radial bowing out from their support brackets as though the elements had grown in length, but were restrained from growing in the lineal end-to-end direction. A new furnace, similar to the failed furnace, but with rods perpendicular to the elements was installed for PG-7 to limit the bowing.

Additional data for the run is being assembled and will be reported in the next quarterly.

INTERMEDIATE AND LOW LEVEL WASTE TREATMENT

MELTER CONDENSATE FROM PHOSPHATE GLASS PROCESS

Development Studies - W. T. McKean

A series of three hot runs have been made distilling actual sulfate containing melter condensate from phosphate glass solidification Run PG-4 in the Distillation Demonstration Unit (DDU). Distillation pressure, rate of heat input, feed volume, and feed and product transfer procedures were similar to earlier cold runs described in previous quarterly reports." Table 5.7 lists the operating conditions for the three runs.

Acid separation efficiencies under these operating conditions were quite good as shown in Figure 5.2 where the distillate concentrations of nitric acid, sulfuric acid, and ruthenium are shown as a function of distillation time. At an elapsed time of 7.3 hr, corresponding to 78% volume distilled, the distillate was switched from the nitric acid receiving tank

*BNWL-1003, 68-4, and BNWL-1074, 69-1.

TABLE 5.7. *Summary of Distillation Demonstration Unit (DDU)
Hot Runs*

Run No.	Boilup, gal/hr	Pressure, mm Hg	DF ^(a)	
			H ₂ SO ₄ fraction	HNO ₃ Fraction
1H	1.8	50	75	27
2H	1.6	50	60	12
3H	1.9	30	220	34

(a) $DF (H_2SO_4 \text{ Fraction}) = \text{Ruthenium in Feed} / \text{Ruthenium in } H_2SO_4 \text{ Fraction}$.
 $DF (HNO_3 \text{ Fraction}) = \text{Ruthenium in Feed} / \text{Ruthenium in } HNO_3 \text{ Fraction}$

FIGURE 5.2. *Sulfuric Acid, Nitric Acid and Ruthenium Concentrations in Distillate as a Function of Time. (Run 2H)*

to the sulfuric acid tank. Approximately 8% of the sulfuric acid charged to the evaporator was collected to the nitric acid tank, 75% in the sulfuric acid tank, and the 17% remained in the evaporator. All nitric acid charged to the evaporator was distilled.

Ruthenium evolution is directly related to the period of maximum nitric acid evolution as shown in Figure 5.2. As residual nitric acid is distilled from the evaporator, the ruthenium evolution subsides. Clearly, the decontamination factor (DF) of the sulfuric acid fraction is dependent on the point of tank switching; in Figure 5.2 a delay in switch point to 8 hours would substantially decrease ruthenium content of the sulfuric acid fraction. Of course, along with this increase in DF of the sulfuric acid stream would go an increase in sulfuric acid content of the nitric acid fraction. Table 5.7 lists the DF values attained in the series of runs. In order to dispose of the sulfuric acid fraction by an asphaltting procedure, it was determined that a DF (H_2SO_4 fraction) of at least 200 would be required based on an activity level of 10 Ci/gal of asphalt. On this basis the DDU concept can be used to decontaminate the sulfuric acid fraction of melter condensate. To attain the necessary DF levels, operating pressures in the range of 30 mm of Hg are required.

INTEGRATED PLANT STUDIES - G. L. Richardson

A study flowsheet that incorporates waste solidification and secondary waste recycle in a conventional Purex reprocessing plant has been developed. The flowsheet segregates condensates from product and waste concentration and acid fractionator products according to the activity level. Relatively clean solutions are recycled to the tail-end of the Purex process for feed makeups and scrub and strip solutions, while the more contaminated acid and condensate streams are used in

feed dissolution and other head-end steps. During normal plant operation, the flowsheet provides more "clean" HNO_3 and condensate than is required in the process, thus providing a convenient method for bleeding excess water and acid from the system.

The bulk of the excess clean acid and water is derived from evaporation and calcination of the UNH product. By definition, the overhead streams from these operations should have only trace amounts of fission products (mostly ruthenium), together with minor amounts of entrained UNH. Final cleanup for disposal, if desired, should be easily accomplished.

Other flowsheet features that favor clean effluent streams include the use of sugar to minimize ruthenium volatilization, use of the aqueous waste stream to scrub nitric oxides and ruthenium from the waste calciner off-gas, and the use of a "fumeless" (O_2) dissolution technique to improve recovery of NO_x from the dissolver and calciner off-gases.

PRODUCT MEASUREMENTS, TESTING AND STORAGE

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

The cubicle radiation shielding problems that were reported last quarter* have been resolved by installation of the original cubicle shielding plugs from inside the cell. Full use of SSETF is expected during the first quarter of fiscal year 1970.

*BNWL-1074, 69-1.

The schedule of WSEP waste containers to be studied in SSETF has been revised to include a PW-2 (PC-6) and a fast reactor (PG-11) solidified waste; the schedule is given in Table 5.8.

Characterization of WSEP Solidified Waste - R. J. Thompson

Figures 5.3 and 5.4 are graphical summaries of effective thermal conductivity data for phosphate glass Runs 1 to 6 and spray solidifier Runs 4 to 10, respectively. A least squares regression analysis was made for the effective thermal conductivity of the solidified waste from each process. The resulting equation is presented with the data for both products. The SS-6 data were not included in the regression analysis for spray solidifier product because of the significantly different thermal history of SS-6. Storage of SS-6 at a high temperature (centerline temperature of 900 to 950 °C) for approximately 24 hr in B-Cell resulted in a significantly higher effective thermal conductivity at high temperatures for the product. The effect of the high temperature storage will be the subject of further study in the SSETF where temperature conditions can be maintained more carefully than in B-Cell.

The thermal conductivity of the products from both processes exhibits moderate temperature dependence. In general, the spray solidifier product has a higher effective thermal conductivity than phosphate glass product. However, at higher average temperatures the effective thermal conductivity of the two products is more nearly the same.

The slopes of the effective thermal conductivity curves for the individual runs do not vary greatly from the slope of the least squares regression line for the two products

TABLE 5.8. SSETF Environmental Test Plan

Test No.	Process (a) Run No.	PW Feed Type	Initial Heat, kw	Pot Material	Storage Media
1	PG-4	2	5.8	MS	Air
2	SS-4	2	5.2	MS	Air
3	PC-6	2	5.1	304L	Air
4	PG-6	1	9.6	304L	Water
5	SS-8	1	6.8	304L	Air
6	SS-9	4m	7.2	310	Water
7	SS-10	4m	7.2	MS	Air
8	PG-7	2	10.8	310	Air
9	PG-8	4m	8.5	MS	Water
10	PG-10	4m	8.5	304L	Air
11	PG-11	Fast (b) Reactor	8.5	304L	Water
12	SS-11	Fast Reactor	9.0	304L	Water
13	SS-12	4m	10.0	304L	Water
14	PC-8	4m	6.0	304L	Air
15	PC-9	4m	6.0	304L	Water
16	PC-12	1	5.1	304L	Water

(a) PC = pot calcine, PG = phosphate glass, SS = spray solidifier
 (b) Waste resulting from the processing of a fast reactor core
 fuel irradiated to 100,000 MWd/tonne at 200 MW/tonne.

FIGURE 5.3. Phosphate Glass Product Effective Thermal Conductivity

FIGURE 5.4. Spray Solidifier Product Effective Thermal Conductivity

except for SS-6. As noted previously, the high temperature storage appears to have materially affected the spray solidifier product thermal conductivity for this run.

The following product characterization measurements were made in B-Cell during the report period.

A. Pot Calorimetry

1. SS-10: 7.13 kW on 5/2/69
2. SS-3: 0.622 kW on 5/7/69
3. SS-5: 5.11 kW on 6/2/69
4. PG-7: 9.55 kW on 7/16/69

B. Afterfill dimensions

1. PG-5: No significant dimension changes noted.
2. SS-10: No significant dimension changes noted.

C. Pot Leak Testing

1. SS-9: 2.1×10^{-7} atm cm³/sec
2. SS-10: 2.5×10^{-7} atm cm³/sec

D. Kiethly Scans

1. PG-6 1.2×10^4 R/hr max (5/12/69)
2. SS-10 2.5×10^4 R/hr max (4/25/69)

E. Temperature Profiles

1. PG-6: air
2. SS-3: H₂O
3. SS-10: air and H₂O
4. SS-5: H₂O
5. PG-7: air and H₂O

6. HEAVY SECTION STEEL TECHNOLOGY PROGRAM

IRRADIATION EFFECT ON THE FRACTURE OF HEAVY SECTION

PRESSURE VESSEL STEELS - C. W. Hunter and J. A. Williams

Specimen Irradiations

Compact tension fracture mechanics specimens (CTS) and tensile specimens of ASTM A 533-B pressure vessel steel were rotated and exchanged in the irradiation assembly during the April outage of the ETR. The initial reactor charging of this assembly into the M-3 loop of the ETR, during Cycle 101 in February 1969, has been previously described.⁽¹⁾ Four CTS and 15 tensile specimens were removed and replaced with unirradiated specimens. The end specimens in the assembly were rotated for irradiation during Cycle 102 to compensate for the flux gradients. The fast fluence of those specimens removed was 1.5 to 2.5×10^{19} nvt (>1 MeV), based on the fission spectrum of the reactor. Additional flux monitors positioned in the grip holes of the CTS are being analyzed. A plot of the fast flux distribution over the length of the irradiation assembly is shown in Figure 6.1.

Figure 6.2 presents the temperature distribution along the assembly length during full reactor power. These temperatures were measured at the center of the root of the machined notch of the CTS. The tensile specimen temperature during full-power operation was 503 °F as measured in the button head at a bulk water temperature of 500 °F, and varied with the bulk water temperature. The typical water temperature was 498 to 503 °F at full reactor power. Temperature rise of the water during its flow through the irradiation facility was negligible. The flow rate was 100 gpm and the pressure was 2000 psi. The temperature gradients, indicated in Figure 6.2 for full-power operation, decreased proportionately as the reactor power and water temperatures decreased. The gradients therefore were compressed and the specimen temperatures

Figure 6.1 Flux Distribution Over the Length of the ETR, M-3 Loop, HSST Specimen Irradiation Assembly

converged at the water temperature during reactor shutdown. The reactor power and M-3 loop water temperature for Cycle 101 are recorded in Figure 6.3.

Figure 6.2 Temperature Distribution over Length of ETR, M-3 Loop Irradiation Assembly. Temperatures Measured at the Center of the Notch Root of IT Compact Tension Specimens. Temperature Distribution Measured at Full Power.

Figure 6.3 Bulk Outlet Water Temperature of M-3 Loop and Reactor Power for ETR Cycle 101

Cycle 102 outage occurred in July, at which time eight CTS and 14 tensile specimens of ASTM A 533-B base material were removed and replaced with eight CTS from the submerged arc weldment and with 14 tensile specimens of plastically strained base material for the strain aging embrittlement study. The four CTS specimens occupying the center position of the assembly since charging during the Cycle 101 outage will remain in position to accumulate a total fluence of 8×10^{19} nvt.

Initial heat transfer studies for the design of an irradiation capsule for 4-in. compact tension specimens (4T CTS) have been completed. The calculations were based on scattered gamma data from possible "equivalent" positions throughout the ETR. However, the results did establish the feasibility limits of capsule irradiation. In addition, some modes of capsule operation to gain a variety of irradiation conditions were suggested by the results. The predicted temperature distributions are shown in Figure 6.4 for three planes normal to the vertical axis of the reactor. These planes correspond to the fracture planes of the three specimens in the capsule. The temperature distribution was calculated for the heat transfer gaps and gamma heat generation gradients shown by using heat transfer coefficients for a mixture of 50% He-50% Ar in the gap. The results indicate that multiple irradiation temperatures could be obtained in the same irradiation capsule and that, for single irradiation temperatures, a varying gas gap over the length of the capsule would be required. Comparison of Plane B for two different gamma heat values represents the extremes on which the gap design must be based. At the higher gamma heat rate, only a 0.002-in. gap is allowable to obtain temperatures in the range desirable. Larger temperature gradients are also exhibited for higher values of gamma heat. A 0.030-in. gap at the high gamma heat rate yielded temperatures exceeding the melting point of the material. In addition, the calculations are based

Figure 6.4 Irradiation Temperature Distribution of 4T Compact Tension Specimens at Fracture Planes. Temperatures were calculated for the gamma heat generation shown over the axial length at the capsule with a mixture of 50% He - 50% Ar in the gas gap.

FIGURE 6.5 Fracture Toughness and Yield Strength as a Function of Temperature for ASTM A 543, Grade B, Class I Steel from HSST Plate 02. Closed points on the yield curve represent 0.2% offset yield strength. Valued closed points on the fracture curve were invalid K_{Ic} determinations, based upon the ASTM Criteria, a or B 2.5 (K_{Ic}). All specimens from the quarter through center positions were for measuring properties in the longitudinal (rolling) directions, i.e. fracture specimens were RW and tensile specimens were R.

unrealistically on constant value of gamma heat over the length of the capsule. However, the results indicate that irradiation of 4T compact tension specimens is feasible if the gamma heating is not too high. Therefore, accurate gamma heat generation experiments for the positions considered for capsule irradiations in the ETR are currently being conducted.

Base Material Studies of HSST ASTM A 533-B

Base material studies of unirradiated HSST material, Plate 02 of A 533-B, conducted during the last quarter, included fracture toughness and tensile testing. The unirradiated specimen testing at PNL on base plate material is oriented toward providing a cursory examination of mechanical properties for comparison with data yielded by other HSST investigations of base plate material.

The measured fracture and tensile properties are shown in Figure 6.5 as a function of test temperature. The specific data are tabulated in Tables 6.1 and 6.2. Both the fracture toughness and tensile data are comparable to that of other investigators of the HSST program. However, some small material differences may be observed in the tensile data. The present values of yield strength reported in Table 6.1 are slightly lower than those reported by ORNL⁽¹⁾ and elsewhere in the same reference by Westinghouse. Total elongation of these data and the Westinghouse data are comparable, while the ORNL material exhibited slightly higher values of total elongation. The variations between the data are most probably due to variations in plate properties.

Table 6.1 Tensile Properties of A 533, Grade B, Class I Steel From HSST Plate 02, Longitudinal Specimens

Specimen Identity	Test Temperature		Plate Position	Yield Strength, $\text{psi} \times 10^3$		Ultimate Strength, $\text{psi} \times 10^3$	Total Elongation, %	Reduction In Area, %
	$^{\circ}\text{F}$	$^{\circ}\text{C}$		Upper	Lower			
02GA 194	-250	-156	7/19	103.8	97.7	119.1	24.3	60.6
02GA 221	-250	-156	9/19	101.2	97.3	127.9	24.5	44.1
02GA 147	-250	-156	5/19	107.6	101.3	136.1	24.2	60.7
02GA 171	-250	-156	6/19	108.2	100.4	132.6	24.6	60.2
02GA 195	-150	-101	7/19	83.7	81.6	108.8	18.0	66.5
02GA 196	-50	-45	7/19	70.8	70.2	93.8	19.6	50.6
02GA 197	75	24	7/19	67.5	65.4	86.5	17.5	68.3
02GA 223	76	24	9/19	67.2	66.4	85.7	17.5	71.0
02GA 198	275	135	7/19			59.7	15.0	35.8
02GA 225	475	246	9/19			57.2	11.6	65.4
02GA 175	475	246	6/19			61.0	15.6	58.9
02GA 199	475	246	7/19			59.4	15.0	63.6
02GA 200	575	300	7/19			55.3	17.4	66.0

* Plate position - fractions of plate thickness at specimen axis.

Table 6.2 *Fracture Toughness, K_{Ic} , of A 533-B Steel from HSST Plate 02, Measured with IT Compact Tension Specimens, RW Orientation at 4/9T Plate Position*

Specimen Identity	Test Temperature		Crack Length a, in.	Yield Strength, psi $\times 10^3$	K_Q ksi $\sqrt{\text{in.}}$	$2.5 \frac{K_Q}{\sigma_{ys}}$	K_{Ic} ksi $\sqrt{\text{in.}}$
	$^{\circ}\text{F}$	$^{\circ}\text{C}$					
02GA 28	-250	-156	0.974	105	31.2	0.220	31.2
02GA 5	-185	-121	1.005	91	39.8	0.478	39.8
02GA 6	-185	-121	1.014	91	43.3	0.566	43.3
02GA 8	-135	-93	1.039	83	41.0	0.610	41.0
02GA 25	-135	-93	0.972	83	49.7	0.896	49.7
02GA 47	-100	-73	0.927	78	53.0	1.518	
02GA 26	-100	-73	0.994	78	68.0	1.900	
02GA 48	-50	-45	0.942	72	60.2	1.74	
02GA 27	-50	-45	0.918	72	68.6	2.26	

Weldment Study of ASTM A533-B

The weldment currently under study is the HSST program weldment section 51A which had been submerged arc welded from HSST program plate 01. The original size was 30 in. long by 12 in. thick. The investigation is utilizing both 1-in. thick compact tension specimens (IT CTS) and 1/2-in. double cantilever beam (DCB) specimens to determine the toughness gradient through the weldment. The CTS are oriented with the fracture plane parallel to the weld and perpendicular to the plate surface, and the notches are at different positions in the weld and HAZ. The DCB specimens are oriented with the fracture plane perpendicular to the weld and plate surface so that the successive propagation events assess the toughness gradient. These specimen orientations are schematically depicted in Figure 6.6.

The metallurgical areas for location of the notches in the CTS are indicated in Figure 6.7. Probably the most important areas are the weldment (A), the weld-dilution region or weld-side of the fusion line (B), and the region of austenitic grain growth or HAZ-side of the fusion line (C). Four IT CTS from Area A and four from Area B were charged into the M-3 loop for irradiation during Cycles 103 and 104 to fluences of $1 \text{ to } 2 \times 10^{19}$ nvt at approximately 540 °F. Between Cycles 103 and 104, four specimens from Area C will be charged into the M-3 loop. Similarly positioned, unirradiated specimens will also be tested.

Results of unirradiated 1/2-in. DCB tests at -256 °F (-160 °C) and -278 °F (-172 °C) are depicted in Figure 6.8. The results are from three specimens at each temperature. The typical dimensions for the specimens were a gross width of 0.500 in., a double-beam height of 1.000 in., a net width of 0.183 in., a notch radius of 0.001 in., and length of 7 inches. All the specimens were from quarter-thickness through center positions. The critical stress intensity at crack initiation and arrest (K_c and K_a) were calculated from the loads and crack

Figure 6.6 Macrograph (1X, 5% Nital Etch) Through Thickness of Submerged Arc Weldment. The CTS and DCB specimens are to be utilized in determining the toughness gradient through the weldment.

Figure 6.7 *Locations of Areas of Interest in Weld Metal and HAZ of Submerged Arc Weldment*

Figure 6.8 Distribution of Critical Fracture Toughness for Initiation and Arrest Through Base, HAZ, and Weld Metal as Determined by 1/2-in. DCB Specimens at -256 and -278 °F. The closed points at -278 °F are all from one specimen which yielded consistently low values.

length using the expression^(2,3)

$$K = \frac{4P^2}{hBB_n} (3x^2 + 1)$$

in which $x = \frac{a}{h} + 0.6$

P = critical load

h = beam height, 0.500 in.

B = gross width, 0.500 in.

B_n = net fracture width, 0.183 in.

a = crack length from point of load application.

The crack lengths were both measured and calculated from the experimental compliance calibration for this specimen determined by Hoagland.⁽⁴⁾

The critical stress intensity (ksi√in.) data for crack initiation and arrest in the base metal and weld are summarized as follows:

Temperature	BASE		WELD	
	K _c	K _a	K _c	K _a
-256 °F	40	34	47	30
-278 °F	36	28	38	27

The principal and significant feature of the data is that the weld metal has a higher initiation toughness but a lower arrest toughness than the base material. Such a behavior indicates the weld metal to be more rate sensitive. At -256 °F, the exclusion of any data in the HAZ is indicative of low toughness in this region. However, the initiation and arrest point in the HAZ at -278 °F are not particularly low. The closed points at -278 °F are all from one specimen. The testing of this specimen during a period of high relative humidity resulted in a badly iced cryostat. This condition probably was responsible for the consistently low values. Limits for valid K_{IC} data are indicated on the right side of Figure 6.8. These data, determined for a yield strength of

11,000 psi, utilized the following criteria:

- $B \geq 2.5 (K_{IC}/\sigma_{ys})^2$, from Reference 5, giving a K_{IC} maximum of $49 \text{ ksi}\sqrt{\text{in}}$;
- $B \geq 4 (K_{IC}/\sigma_{ys})^2$, from Reference 6, giving a K_{IC} maximum of $39 \text{ ksi}\sqrt{\text{in}}$.

Strain Aging Embrittlement

The intent of this study is to determine the deleterious effects of irradiation and thermal aging upon a vessel subjected to plastic strain at the roots of cracks, flaws, or notches. Such plastic strain will result from both the original hydro-proof test and from normal service operation. This present study is relevant to vessel integrity by simulating in precracked specimens the above conditions.

Strain aging effects are pertinent to fracture initiation rather than to propagation and arrest because, while plastic strains and blunting occur only at the crack tips, they may continually degrade fatigue crack resistance. Also, the strain and blunting conditions cannot be identically reproduced in a bulk material. Thus these effects have meaning within the realm of fracture mechanics, and one may expect the simulation of the load conditions in a precracked specimen to accurately predict vessel behavior.

A diagrammatic representation of the experimental work is shown in Figure 6.9. The material is the same as that utilized in the base metal study, HSST Program plate 02GA. The fracture specimens are IT CTS from 1/4 through 3/4 thickness positions oriented in the RW direction. After fatigue crack preparation, the stress intensities are applied to strain the metal at the crack root. After irradiation or thermal aging, the extent of embrittlement will be assessed by comparing the K_{IC} versus temperature behavior of these specimens with the behavior of unaged specimens.

The temperatures of stress intensity applications correspond to the typical proof test and operating temperatures. The stress intensities are within the range of operating and proof test belt-line stress levels on elliptical surface flaws 0.5 to 1.0 in. deep.

This present strain aging study may be differentiated from warm prestressing on the basis that the latter supposedly is to produce beneficial effects by crack blunting in an irradiation embrittled material. The study of warm prestressing will not be started until the effects of strain aging embrittlement are known.

Presently, 15 precracked specimens have been strained under the conditions described in Table 6.3. The extent of plastic deformation occurring at the crack tip is just as important as the stress intensity level and temperature. Therefore, the notch opening was measured at the front of the specimen to determine the K-level at which deviation from linear elastic behavior and the extent of permanent notch opening displacement would occur. The precision of the strain gage extensometer used at 100 °F was better than 0.0001 in., while that of the LVDT used at 550 °F was about 0.001 to 0.002 inch. At 100 °F, below a K-level of $40,000 \text{ psi}\sqrt{\text{in.}}$, linear elastic behavior did not deviate. The indicated plastic displacement occurred in the 1-hr hold only during the 40,000 to 60,000 psi loading. At 550 °F, the load-displacement record indicated no appreciably greater deviation from linearity during loading than for the 100 °F case, but that additional plastic displacement occurred during the hold. These specimens will be thermally aged at 550 °F for about 30 days and then tested.

Tensile specimens have also been plastically strained and are being irradiated and thermally aged to assess the effects of strain aging embrittlement on flow properties. This information will permit a more lucid analysis of the strained metal at the

crack tip in the fracture specimens. The tensile specimens have been plastically strained 1% and 10% at 100 °F and seven specimens from each condition were charged into the M-3 loop for irradiation during Cycles 103 and 104.

STRAIN AGING INITIAL STUDY

Figure 6.9 Diagrammatic Representation of Study to Determine Extent of Embrittlement in Plastic Strain Region at a Crack Tip Produced by Subsequent Neutron and Thermal Aging

*Table 6.3 Conditions of Stress Intensity Application to
Precracked IT Compact Tension Specimens*

Specimen No.	Plate Position	K-Level psi/in.	Temperature, °F	Crack Length, in.	Load, lb	Plastic Displacement, in.
02GA 13	1/2-3/4	35,000	100	1.041	4839	<0.0001
02GA 15	1/2-3/4	35,000	100	1.038	4867	<0.0001
02GA 29	1/2	35,000	100	0.999	5155	<0.0001
02GA 35	1/2-3/4	35,000	100	1.035	4876	<0.0001
02GA 56	1/2-3/4	35,000	100	1.007	5097	<0.0001
02GA 14	1/2-3/4	60,000	100	1.039	8321	0.0004
02GA 33	1/2-3/4	60,000	100	0.987	9008	0.0007
02GA 30	1/2	60,000	100	1.033	8408	0.0010
02GA 34	1/2-3/4	60,000	100	1.023	8518	0.0005
02GA 36	1/2-3/4	60,000	100	1.015	8648	0.0007
02GA 53	1/2-3/4	35,000	550	1.015	5029	0.001*
02GA 31	1/2	35,000	550	0.971	5379	0.001*
02GA 54	1/2-3/4	60,000	550	0.951	9479	0.006*
02GA 55	1/2-3/4	60,000	550	0.972	9980	0.002*
02GA 32	1/2	60,000	550	1.017	8613	0.002*

* *Extensometry at 550 °F for the 1-hr hold time was not sufficiently reliable to provide a more accurate plastic displacement measurement.*

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