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**NUCLEAR SAFETY QUARTERLY REPORT
FEBRUARY, MARCH, APRIL, 1968**

**FOR
NUCLEAR SAFETY BRANCH
OF
USAEC DIVISION OF REACTOR
DEVELOPMENT AND TECHNOLOGY**

OCTOBER 1968

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By
The Staff of Battelle-Northwest

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FOREWORD

This report is the fifth of a series in which Pacific Northwest Laboratory reports its Nuclear Safety-related studies being performed for the USAEC Division of Reactor Development and Technology. Previous reports in this series are:

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

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1. SUMMARY

ENGINEERED SAFEGUARD STUDIES

Containment Systems Experiment (CSE)

CSE construction was continued with installation of a 1 in. layer of thermal insulation on the CSE containment vessel, the installation of equipment needed for planned leakage rate experiments with heated air and steam-air containment atmospheres, and the fabrication of additional penetrations in the vessel.

Supporting development work on methods for generating aerosols at a linear rate for extended periods of time was performed. Techniques were demonstrated for release of a cesium aerosol at a linear rate of 3.5 g/min during a 2.5 hr release period. The method involves metered injection of cesium hydroxide solution into a vaporization tube held at 1200 °C in a furnace. An air sweep gas carries the cesium aerosol from the furnace tube through a heated, 20 ft long delivery line to the point of release.

Computer programs were written to prepare summary tables and machine plotted curves of the on-line stored gamma energy analytical data from the fission product transport experiments. Plotting programs for blowdown data were also completed, and data from Runs B-15 through B-18 were made available as machine plotted figures.

One of the thrust supports for the reactor simulator vessel failed during Run B-18 allowing the vessel to move rearward until it was stopped by surrounding structural support. Extensive nondestructive tests were performed on the vessel following the accident. Ultrasonic tests revealed loss of back reflection in an extensive area and scattered depth indications at about 1.5 in. from the outer surface of the bottom head near

the blowdown nozzle. In addition, some enlargement of a previously noted defect indication in the top head was noted. No defect indications were found by radiographic, dye penetrant, and magnetic particle tests of all areas of the vessel subject to high stresses during blowdown service or the accident.

Extensive preliminary calculations were made to analyze the stresses due to mechanical and thermal loads on the reactor simulator vessel. The objectives were to attempt to determine whether the blowdown service or the thrust support failure accident could account for the present laminar defect indication, and to determine the potential failure modes if substantial further growth of the defect indication should occur. The principal conclusions were that none of the stresses calculated were high enough to initiate a failure in sound material. However, assuming the presence of a plane of defective material, the thermal stresses estimated from blowdown service or transient stresses during failure could be sufficiently high, under some stress intensification, to cause the defect. The most probable failure mode appeared to be associated with propagation of the defect plane into and around the blowdown nozzle bore.

Fission Product Aerosol Control

Additional spray demonstration runs were performed in a steam-air mixture in the pressurized aerosol tank. With 10% hydrazine, the removal half-time was 7 to 8 min at a spray rate of 0.08 gpm/ft. Wall washing was found to account for about half the removal, and volume for volume was 2 to 3 times as effective as the droplets.

Reaction rates between hydrazine and methyl iodide in the liquid system were extended to higher concentrations and temperatures. The improved method gave good reproducibility. The role of hydrolysis in depleting methyl iodide was found to

be of little importance to hydrazine solutions in concentrations of practical study interest.

PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

Crack Detection in Pressure Piping by Acoustic Emission

Investigation of the effects of the transmission media and specimen geometry on propagation of acoustic emission signals is in progress. Evidence indicates that there is a short duration packet of information at the leading edge of the detected acoustic emission signal which is not affected by specimen geometry, but is, rather, characteristic of the source mechanism. It also appears that selected mode detection, i.e., shear wave or Rayleigh wave by critical angle selection, would be advantageous to signal detection and to source location functions.

The PZT-5A transducer assembly used for acoustic emission study work was modified to achieve about a five-fold increase in sensitivity. A high temperature (600 °F) life test of this assembly is in progress with no indication of crystal deterioration after 500 hr of continuous testing. A lithium niobate piezoelectric transducer with improved sensitivity is being assembled for testing at elevated temperature (>700 °F).

A program prepared jointly with Phillips Petroleum Co., has been directed toward developing three successive levels of refinement in acoustic emission monitor systems for integrity surveillance of reactor pressure boundaries. In general, PNL is responsible for R&D work and PPCo. is responsible for plant application test work under this program.

Acoustic emission data obtained during burst tests of two 24 in. diam, 1.64 in. wall, type A-106-B steel pipe specimens at the Battelle-Columbus test site have been analyzed. Both tests involved part-through-wall flaws. Low level signals

attributed to metal deformation controlled the shape of the emission rate curve. The curve showed a distinctive preliminary peak in both tests about 4 min prior to failure. A small amount of slow crack growth was indicated in both specimens. Burst signals signifying this growth contained a unique packet of high frequency energy at the leading edge which appeared to be independent of geometry effects.

Arrangements have been made to install instrumentation on Commonwealth Edison's Dresden 1 nuclear power reactor to examine the background noise spectrum during startup and operation.

FIXATION OF RADIOACTIVE RESIDUES

During the report period, approximately 3×10^6 Ci of radioactivity were successfully processed in two phosphate glass runs in WSEP. These runs represent the fourth and fifth conversion of highly radioactive wastes into glass on an engineering scale. Phosphate glass Run PG-4 was made using sulfate-containing Purex Waste 2 (PW-2) feed, while PG-5 was made using sulfate-free Purex Waste 1 (PW-1) feed. Leach rates on phosphate glass solids from WSEP were 1×10^{-6} to 2×10^{-7} g/cm² x day.

Platinum corrosion was evaluated for sulfate-free phosphate glass flowsheets made with excessive phosphate, lead, and silicone antifoam agent. Significant platinum corrosion occurred only in the vapor phase with high phosphate feed, and in the melt phase with feed containing lead. In no case was there evidence of cracking or embrittlement.

Testing of low-salt flowsheets was carried out on the developmental spray solidifier. Major characteristics were determined to be reduced capacity in the calciner, reduced denitration of the powder from spray calciner, and increased difficulty in removing the calcined powder from the spray calciner filters.

A simulated spray-solidified melt prepared from low-salt PW-4 solution corroded both 304L and 310 SS at a rate of about 15 mm/month in 100-hr tests at 800 °C.

Addition of formaldehyde or hypophosphorous acid to spray solidifier feed was found to reduce the volatility of ruthenium significantly.

Laboratory tests with commercial silicone antifoam agents were successful in reducing DBP-induced foaming in phosphate glass runs.

Measurements on grab samples of phosphate glass product showed leach rates of 10^{-6} to 10^{-7} g/cm²/day.

Hypophosphorous acid addition to spray solidifier feed resulted in markedly less ruthenium volatility. Formaldehyde additions were less satisfactory.

2. ENGINEERED SAFEGUARD STUDIES

CONTAINMENT SYSTEMS EXPERIMENT - G. J. Rogers

Modifications to Experimental Facilities - C. E. Linderoth

Following the interruption in December, 1967, of experimental activities involving the containment vessel, work was continued by on-site construction forces to insulate the containment vessel, install air-heating and recirculation equipment inside the containment vessel, install more penetrations, and modify several existing penetrations on the vessel.

The entire containment vessel surface was covered with 1 in. thick Fiberglas (6 lb/ft^3 , $k = 0.026 \text{ Btu/(hr) (ft}^2) (\text{°F/ft})$) insulation. On the outside surface of the insulation is a layer of 0.0025 in. thick, factory applied aluminum. The insulation is held in place by spring washers slipped over 4 1/4-in. long pins, spot welded to the vessel. The technique for attaching the pins was in accordance with Section VIII of the ASME Code for Unfired Pressure Vessels. Excessively long pins were used for attachment in order to permit the future application of two additional layers of insulation.

The equipment installed to heat the vessel atmosphere for the heated air leakage rate and the heated steam-air leakage rate experiments is schematically shown in Figure 2.1. The gas to be heated is drawn from the bottom of the containment vessel through a thin walled duct to a finned tube heat exchanger. Both steam and cold water were piped to the heat exchanger for use in heating or cooling the gases. From the heat exchanger, the gases pass through a 4000 scfm fan and are discharged via a vertical stack made of light gage sheet metal. A draft tube is incorporated in the stack to mix cooler gases with the gases from the heat exchanger and thus

FIGURE 2.1. *Equipment for Leakage Rate Experiments with Heated Air Atmosphere*

minimize extreme thermal gradients in the vessel dome. After the leakage rate experiments are completed, this equipment will be removed and the fan and heat exchanger will be saved for experimentally determining the effectiveness of air cleaning systems.

Ten penetrations, all of the pipe flange type, were installed or modified on the containment vessel in accordance with Section VIII of the ASME Code for Unfired Pressure Vessels. Early experiments showed that the pools of water which formed in the bottom of the containment vessel and dry-well were not adequately mixed prior to sampling and, consequently, the uncertainties of the true chemical concentrations in these pools were undesirably high. Three penetrations were modified (two on the containment vessel and one on the dry-well) as return lines for the pool water drawn from the bottom of the vessels through appropriately sized pumps. Eductors are to be installed at the discharge points to promote further the mixing of the water. Similar wet well mixing problems could very well arise when pressure suppression type fission product transport and behavior studies are performed. In preparing for such problems, an additional penetration was added on each of the four wet wells. These penetrations until they are needed for water recirculation, will be available for thief sampling the aerosols in these compartments. Three penetrations were installed on the containment vessel for purposes of serving as either viewing windows or for thief sampling of the aerosol. Two of these were installed above the main deck at the +5 and +15 ft levels. The third was installed at the -15 ft level to permit access into the compartment between the wet wells.

Extended Linear Rate Release of Cesium Aerosols - L. F. Coleman

High temperature vaporization of cesium hydroxide solutions is being investigated as a technique for releasing cesium aerosols at linear rates of from 1 to 10 g/min.

Long duration linear release rates of aerosols will be needed for validating mathematical models for aerosol transport and behavior in containment. In this specific case, we wish to demonstrate techniques for releasing up to 5 g of cesium per minute for durations of 2 hr with a linearity better than $\pm 20\%$. Physical properties of the aerosol also must be characterized at experimental conditions enabling relation of the aerosol dynamics to previous burst type, low mass cesium release experiments performed in CSE.

Figure 2.2 shows the cesium aerosol generation apparatus under evaluation. The apparatus consists of a silicon carbide resistance furnace (operated at 1200 °C), a 1 in. diam nickel tube packed with nickel turnings and a water cooled, stainless steel injection tube. A worm gear driven set of infusion-withdrawal syringes serves as the linear solution metering device. An inert, immiscible fluid (silicone oil) is used as the pumping fluid so that the pumping apparatus will remain free of contamination and can be located outside the radiochemical hot cell to conserve cell space and minimize remote control requirements.

Linearity of cesium release with time is illustrated for this method in Figure 2.3 where a release rate of 3.5 g/min was obtained at the exit end of a 20 ft long, 2 in. diam aerosol transit line heated to 150 °C. Input rate to the delivery line was 7.3 g/min for a yield of 48% with 51% plate out in transit.

Solution feed rate was 5 ml/min (1.4 g of cesium per milliliter, cesium hydroxide solution) into the 1200 °C vaporized tube. At the sweep gas flow of 0.5 scfm, the gas loading was 515 g Cs/m³ and 248 g Cs/m³ at entrance and exit of the 20 ft aerosol delivery line, respectively.

FIGURE 2.2. *Apparatus for Sustained Linear Release of Cesium Aerosol*

FIGURE 2.3. *Cesium Aerosol Generation Rate by CsOH Solution Evaporation*

Particle size analysis of the aerosol was obtained by inertial impaction methods. Mass median diameters (MMD) of 4 to 5 μ were determined for the aerosol at entrance and exit ends of the 20 ft delivery line. Previous tests at cesium generation rates of 1 to 4 g/ml gave mass median particle diameters between 0.8 and 2.5 μ . These values of the mass median particle diameters are greater than that observed in an actual containment test (CSE A4, mass median cesium aerosol diameter of 0.86 μ and cesium concentration during generation of 10 g/m³), possibly due in part to the much higher mass concentration present in the high mass linear rate release tests. A continued study of the effects of solution concentration, feed rate, sweep gas flow rate evaporation temperature, and containment environmental conditions will serve to more fully define the particle size characteristics of cesium aerosols generated by the cesium hydroxide volatilization technique.

Aerosol Data Reduction - L. F. Coleman

Additional computer programs have been written to transfer, organize, and machine plot on-line stored gamma energy analytical data from the fission product transport experiments at CSE. Gamma energy analytical data is first accumulated at the CSE site with the aid of a small digital computer.^(1,2,3) These magnetic tape-stored data are thereafter transferred to a second magnetic tape compatible with a Univac 1108. Tabulated output from the 1108 program includes aerosol concentrations versus time for all sampling locations, errors, fractional amounts of a given radionuclide on various components of a sampler system, mean values, standard deviations, corrections for background activity, and correction for elemental iodine sorption of filter components of Maypack aerosol samplers. Selected data from these tabulated results can also be machine plotted at the time of output.

This machine plotting capability will greatly reduce the time and labor involved in preparing figures illustrating the results of the CSE fission product transport experiments.

The general purpose plotting package can:

- Produce up to 15 curves per plot plus standard deviations
- Scale X and Y axes
- Produce linear or semilog plots, with or without guide
- Point plot, line plot, or fair curve
- Furnish multiple titles on both axes.

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COOLANT BLOWDOWN STUDIES - R. T. Allemann, A. J. McElfresh, W. C. Townsend, M. E. Witherspoon, and N. P. Wilburn

Experimental Blowdown Results

Continued analysis of the blowdowns conducted in previous quarters was delayed while computer program printout and plotting refinements were made. Experiments had been conducted in a large 150 ft³ reactor simulator vessel (Figure 2.4) mounted in a test stand as shown in Figure 2.5. A series of subcooled blowdowns with starting conditions shown in Table 2.I was made.

The computer plotting routine programs have been developed and results showing some of the primary data for blowdown

FIGURE 2.4. CSE Reactor Simulator Vessel

Neg 0682404-7

FIGURE 2.5. CSE Blowdown Facility

TABLE 2.1. *Subcooled Blowdown Series*

Run No.	Pressure, psia	Weight of Liquid, lb	Temp., °F	Nominal Diameter, in. and Break Area, ft ²
B-14A	2160	7400	500	4 (0.0643)
B-14B	2160	7275	512	4
B-15	2185	6700	560	4
B-16A	2384	6000	600	4
B-16B	2385	6330	590	4
B-17	2375	6300	610	2 (0.0156)
B-18	2400	6350	600	8 (0.253)

Run B-15 are shown in Figures 2.6 through 2.23. These data were taken by a digitally controlled multiplexing system which samples the data sensors at preset frequencies.^(1,2)

Data sampling rates in the early part of the blowdown were set at 500 Hz. After a few seconds (10 to 20 depending on the break area) a slower rate of 200 to 100 Hz was begun. This change of rate sometimes shows up as a change in the level of the "hash" appearing in the plot (Figures 2.9 and 2.11). This hash can be filtered out to give a smoother plot such as would be seen on an analog chart recorder (Figures 2.9 and 2.10). The "5 Hz" in the title refers to the nominal filter cut-off frequency. Figure 2.15 shows that random signals such as occur with the liquid level or with the neutron densitometer are not as effectively filtered, and oscillation in the data comes through the filter.

The data acquisition system, with a sampling rate capability of 5000 Hz, permits study of the extremely rapid decompression of a subcooled liquid. The plotting routine for this data is not yet functioning. The tabulated output voltages have been sufficiently studied, however, to show decompression pressure oscillations to correspond to the acoustic length of the outlet pipe, and that the rupture disk breaking time for a 4 in. break is about 0.56 msec.

FIGURE 2.6. Run B-15 CSE Blowdown (15-10) Pressure
0.125 ft. from Exit (P-8)

FIGURE 2.7. Run B-15 CSE Blowdown (15-9) Pressure
1.08 ft from Exit (P-4)

FIGURE 2.8. Run B-15 CSE Blowdown (15-13) Pressure
1.41 ft from Exit (P-17)

FIGURE 2.9. Run B-15 CSE Blowdown (15-7) Pressure
2.08 ft from Exit (P-3)

FIGURE 2.10. Run B-15 CSE Blowdown (15-7F/5 Hz)
Pressure 2.08 ft from Exit (P-3)

FIGURE 2.11. Run B-15 CSE Blowdown (15-6)
Pressure at Nozzle J (P-2)

FIGURE 2.12. Run B-15 CSE Blowdown (15-8)
Pressure at Nozzle H (P-7)

FIGURE 2.13. Run B-15 CSE Blowdown (15-12)
Pressure at Nozzle R (P-6)

FIGURE 2.14. Run B-15 CSE Blowdown (15-11)
Pressure at Inst. Ring (P-15)

FIGURE 2.15. Run B-15 CSE Blowdown (15-14F/5.0
Hz) Water Level in Vessel

FIGURE 2.16. Run B-15 CSE Blowdown (15-15)
Densitometer History

FIGURE 2.17. Run B-15 CSE Blowdown (15-16)
Weight of Fluid in Vessel

FIGURE 2.18. Run B-15 CSE Blowdown (15-16F/2.5 Hz)

FIGURE 2.19. Run B-15 CSE Blowdown (15-17) Nozzle Thrust Force from LC-2, LC-4

FIGURE 2.20. Run B-15 CSE Blowdown (15-17F/2.5 Hz)
Nozzle Thrust Force from LC-2, LC-4

FIGURE 2.21. Run B-15 CSE Blowdown (15-18) Strain-
gage (S3) Elbow, (S4) Vessel Hoop

FIGURE 2.22. Run B-15 CSE Blowdown (15-25) Temperature 1.25 ft from Exit (T-13)

FIGURE 2.23. Run B-15 CSE Blowdown (15-19) Temperature Nozzle J (T-2)

The pressure data of Figures 2.6 through 2.14 show that the large pressure reduction occurring immediately after the break in the 3.43 in. diam orifice (Figure 2.6) and in the 6.8 in. diam pipe (Figures 2.7 through 2.10) is greatly attenuated in the 41.4 in. diam vessel (Figures 2.11 through 2.14). Subsequent pressures during two-phase blowdown are virtually the same except at the orifice where a large pressure drop results for choked conditions.

Data representing the liquid level in the vessel shown in Figure 2.15 were taken by a time domain reflectometer probe.⁽³⁾ The quite high random noise in this signal is believed to result from flashing of the fluid in the vessel and from the random effect of bubbles on the dielectric constant of the fluid near the probe. Despite these random effects, a mean liquid level declining at an increasing rate and reaching the outlet level at the time of transition can be discerned. The subsequent increase in indication is probably caused by the large change in temperature (after transition) of the expanding steam in the vessel.

The neutron densitometer output for all four detectors is shown in Figure 2.16. The densitometer monitors the effect of the flowing fluid on 14 MeV neutrons, produced just outside the 6.8 in. diam nozzle extension pipe. Calibration and correction curves for the densitometer are needed to determine the void fraction.

Detector D-1 monitors the general neutron flux level which can be used to normalize the data from other channels. The detectors D-2 and D-3 are slow neutron detectors and measure the moderation effect of the flowing mixture. If steam is present, less moderation occurs. Detector D-4 monitors the fast neutrons passing directly through the pipe and absorbed or scattered by the water. This effect, in an opposite sense

to that of moderation and not linear, still gives a measure of void fraction in the pipe. The data in Figure 2.16 show clearly the sharp change in steam-water density at transition.

The weight of fluid in the vessel was deduced from a load cell output. The impulse at the start of the blowdown starts the system into violent oscillations which obscure the data as can be seen in Figure 2.17. Filtering improved the result (Figure 2.18). The filtered data still contains some vibrations, possibly caused by the large oscillations penetrating the cut-off band or representing harmonics of the complex structure holding the vessel. Further refinement of this data will be required before accurate flow rates can be determined. Weight data taken in the past on a chart recorder, while showing the basic oscillation, was greatly smoothed.

The horizontal reaction force resulting from the fluid discharge was measured by two load cells (Figure 2.19). Since the line of action for the discharge is below both load cells, the actual thrust force is determined by taking the difference of the two load cell readings, or by a moment calculation. Filtering of the load cell output also improves legibility of this output (Figure 2.20). The pulse at 10 sec on the LC-4 (load cell-4) filtered output results from the change in data sampling frequency.

The hoop stress strain gage and nozzle-vessel junction strain gage data shown in Figure 2.21 indicate moderate stresses proportional to the internal pressures. No special strains are shown by the gage at the junction of the blow-down nozzle and the vessel.

The temperature of the fluid in the vessel was measured by a series of highspeed thermocouples located at several places in the vessel and outlet pipe. The data from these

sensors (Figures 2.22 through 2.27 show similar temperatures throughout the vessel even when the bulk liquid is no longer touching the upper sensors. The temperature and pressure data correspond to saturation conditions within the accuracy of the data. The temperature sensor in the bottom of the outlet pipe (Figure 2.22) shows the liquid to have been highly subcooled in this region before blowdown. This low temperature has a pronounced effect on the frequency of the subcooled decompression-phase oscillations because of the temperature effect on acoustic velocity.

The cause of the temperature dip occurring near the top of the vessel (Figure 2.26) is not known. This dip, believed to be real because of similar effects seen in other runs is not attributed to sensor malfunction. It may be due to a subcooled condition caused by expansion of the gas in the steam chamber above the liquid before the moisture begins to condense and restore the region to a saturated condition. Note that this is not the same temperature dip accompanying the pressure undershoot observed in LOFT semi-scale blowdown experiments. That dip is also seen, but at the beginning of the blowdown (Figure 2.26). Figure 2.28 shows the temperature in a block of metal fitted in Nozzle "O" to simulate the vessel wall. The thermocouple is separated from the fluid by 3 5/8 in. of carbon steel and 1/8 in. of stainless steel overlay.

The time zero shown on these figures does not correspond to the beginning of blowdown but to the start of the computer sampling. The actual blowdown start, offset from this "time zero" is evident from the pressure and force plots.

FIGURE 2.24. Run B-15 CSE Blowdown (15-20) Temperature Nozzle H (T-7)

FIGURE 2.25. Run B-15 CSE Blowdown (15-21) Temperature Nozzle R (T-6)

FIGURE 2.26. Run B-15 CSE Blowdown (15-22) Temperature Instr. Ring (T-8)

FIGURE 2.27. Run B-15 CSE Blowdown (15-23) Temperature Nozzle O Fluid (T-9)

FIGURE 2.28. Run B-15 CSE Blowdown (15-24) Temperature
Nozzle O Metal (T-11)

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Thrust Support Failure and Vessel Examination - G. J. Rogers

Run B-18 involved an 8 in. bottom nozzle blowdown of coolant from initial conditions of 600 °F and 2400 psig. Failure of the lower thrust support structure during this run allowed the reactor simulator vessel to move rearward until stopped by impact against the surrounding structural supports.

The thrust support failure was the result of an undetected design error during modification of the support structure originally provided.

No visible damage to the reactor simulator vessel resulted from this accident. However, as a precautionary measure, it was decided to perform extensive nondestructive tests on all portions of the vessel subject to large forces or stresses either during the previous blowdown tests or the accident.

No defect indications were found by the radiographic, dye penetrant, or magnetic particle tests of the areas around the 8 in. blowdown nozzles, support trunnions, and vessel leg supports. Longitudinal wave ultrasonic tests revealed an extensive area of back reflection loss, and scattered depth indications at about 1.5 in. from the outside surface in the bottom head of the vessel along an arc below the blowdown nozzle. Some enlargement of a previously noted defect indication was also found on the top head. These loss of reflection areas are considered to indicate laminar defects.

Stress Analysis of the CSE Nozzle C Pressure Vessel

Intersection Area - V. B. Watwood, Jr., and F. R. Vollert

A study was made by the Analytical Mechanics Unit (1) to analyze thermal and mechanical loads influencing past or future growth of the laminar defect, and (2) to determine the failure modes likely to occur with growth of the defect.

The results presented herein should be considered preliminary since some approximate methods were used. However, enough detail and variety of loading conditions have been evaluated to give a fair idea of the dominant nozzle behavior.

All results (with the exception of the failure mode) were based on a geometry ignoring the presence of the laminar defect. This assumption is valid for investigating the stress

fields which might have led to the defect formation. However, with regard to the influence of stress fields on the future behavior of the defect, these results can be used only as an indication. An imperfection will result in a local intensification if the stress field in question has a non-zero stress vector across the failure plane. However, for the case considered here, the normal stress component across the failure plane must be tensile to result in intensification, since the defect faces are assumed to be in contact. It is these types of stress components only that give rise to propagation of the defect. "Turning" of the defect is, of course, a possibility and will be discussed later.

Several loading conditions will now be considered in detail and, in light of the preceding comments, emphasis will be placed on the shear and normal stresses across the defect failure plane. The plane appears to be approximately 1.5 in. in from the outer surface of the shell located reasonably close to the middle surface. Since the shear stress is maximum at the middle surface, the middle surface stress values will be used. For completeness, the maximum combined bending and membrane stresses at the inner or outer fibers will be given. One other item of particular interest is the maximum tensile stress component parallel to the nozzle middle surface in the region where the defect plane intersects the nozzle opening. This stress condition is important because of its relationship to the likelihood of a "weakened plane" projecting into the nozzle opening.

Almost all of the calculations were done with the aid of the computer code, SEAL-SHELL 2.⁽¹⁾ This code is a finite element scheme employing a fairly sophisticated thick shell of revolution element simulating axisymmetric shell structures. It is, of course, subject to those limitations inherent to thick shell theory and is restricted to axisymmetric loading situations.

Nozzle C was assumed to be far enough away from the sphere-cylinder intersection that perturbations arising from that source would be negligible. This assumption, while perhaps questionable, is somewhat more tolerable inasmuch as the defect is on the lower side. This step permits approximating the structure as a nozzle intersecting a half sphere.

The geometrical simulation is shown in Figure 2.29. The largest and most important deviation in the geometry is seen to be in an apparent "thinning out" of the shell in the vicinity of the nozzle-vessel intersection. This deviation should have little effect on the predicted stresses except for the circumferential (hoop) stresses in the vicinity of the intersection and in high predicted values.

The laminar defect was estimated to lie in the region between the fifth and twelfth nodes on the bottom side of the nozzle. Note that the coordinate system in which these stress components are expressed has its x axis along the shell middle surface and the y axis points out of the shell normal to the middle surface. The " θ " axis completes the triad, i.e., σ_{θ} is a hoop stress.

Stresses During Pressurization

The maximum pressure was assumed to be 2400 psi and the elastic constants for a temperature of 600 °F were used. This pressure results in a nominal membrane stress of 6400 psi in the spherical portion of the shell.

However, the presence of the opening and the nozzle walls give rise to local perturbations at the nozzle-sphere intersection. The details of this stress condition were not computed in the original stress report,⁽²⁾ because as stated in the ASME Boiler and Pressure Vessel Code, Section III, Nuclear Vessels,

FIGURE 2.29. Cross Section of Nozzle C as Used in Seal Shell Analysis

compliance with code reinforcement requirements makes it unnecessary. However, since shear and normal stresses on the middle surface could be significant with respect to a defect or weakened plane, their computation was considered worthwhile.

The results are shown in Figure 2.30. The stress indicated by $2 \tau_{\max}$ is the maximum of $\sigma_1 - \sigma_2$, $\sigma_2 - \sigma_3$, $\sigma_3 - \sigma_1$, where the σ_i are the principal stresses.

Finally, note that these stresses are independent of whether Nozzle C is the blowdown nozzle, and that they have existed in the past since the 2400 psi pressure was attained.

Stresses During the Blowdown Transient

If Nozzle C is not the blowdown nozzle, then the pressurization stresses will decrease in direct proportion to the decreasing pressure until such time as thermal stresses come into play. However, if Nozzle C is the blowdown nozzle, then an immediate change in stresses is seen as a result of the pressure drop within the nozzle and in the vicinity of the nozzle opening. Although the pressure drops very rapidly to the saturation pressure (about 1550 psi), the full 2400 psi is applied away from the opening to simulate the worst condition.

Another load source is that imposed on the nozzle elbow by the flowing fluid. Finally, an oscillating tension-compression force will arise in the nozzle due to the sudden release of the initial tension field.

The effect of each of these loads separately and in their most adverse combination is given in Figure 2.31. The moment load on the nozzle cannot be analyzed using SEAL-SHELL 2. Therefore, the methods of Reference (3) were employed to obtain estimates of the maximum bending and membrane stresses.

MAXIMUM COMBINED BENDING AND MEMBRANE
STRESSES - NODES 5-17

STRESS	VALUE (psi)	LOCATION
σ_x	10,500	NODE 11, OUTER FIBERS
σ_θ	14,900	NODE 13, INNER FIBERS
$2\tau_{\max}$	16,800	NODE 13, INNER FIBERS
TENSILE σ_x	950	NODES 11-15 INNER FIBERS

FIGURE 2.30. Stresses Due to Internal Pressure (2400 psig) Prior to Blowdown

MAXIMUM COMBINED BENDING AND MEMBRANE STRESSES,
NODES 5-17

LOADING	STRESS	VALUE (psi)	LOCATION	
INTERNAL PRESSURE }	σ_x	6,000	NODE 10	OUTER FIBERS
	σ_θ	12,000	NODE 12	INNER FIBERS
NOZZLE FLUID }	σ_x	$\pm 14,500$	NODE 12	} OUTER FIBERS +TOP OF NOZZLE -BOTTOM
	σ_θ	$\pm 7,700$	NODE 12	
AXIAL }	σ_x	$\pm 3,400$	NODE 12	OUTER FIBERS
	σ_θ	$\pm 1,800$	NODE 12	OUTER FIBERS

FIGURE 2.31. Stresses During Blowdown Transient (2400 psig Internal Pressure, 8 in. Nominal Nozzle)

These methods do not, however, provide a means of estimating the shear and normal stresses on the middle surface. Therefore, a proportioning procedure was used to make these estimates based on the predicted mid-plane normal and shear stresses given by SEAL-SHELL 2 for an axial loading.

The nozzle has been subjected to stresses of the type just described. The stresses of Figure 2.31 can again be expected with use of the lower nozzle.

Additional Transient Stresses During the Accident

At the time of the accident, the nozzle-vessel intersection was subjected to some additional loads due to inertia of the nozzle assembly. Large accelerations of the nozzle assembly were experienced after failure of the thrust assembly and during the subsequent vessel movement and impact. These loads were estimated and the resulting stresses, combined with the normal transient stresses described in the preceding paragraphs, are shown in Figure 2.32.

Thermal Stresses

Three thermal situations considered to cover the range of the most adverse thermal conditions were investigated. In the selection of these conditions, heavy use was made of the thermal data and comments given in Reference (2).

The first of these thermal situations concerns temperature gradients set up in the nozzle during cooldown after a partial blowdown is performed. The partial blowdown is the more critical since the lower nozzles (e.g., Nozzle C) at an initial temperature of 600 °F remain in contact with 200 °F water until equilibrium is achieved. Reference (2) found the gradient at 3.6 min after blowdown to be the critical one for Nozzle A (only 8 in. nozzle checked) and these results were used herein.

MAXIMUM COMBINED BENDING AND MEMBRANE STRESSES,
NODES 5-17

LOADING	STRESS	VALUE (psi)	LOCATION
ACCIDENT DYNAMIC LOADS	σ_x	$\pm 22,200$	ALL ABOUT NODE 12
	σ_θ	$\pm 11,800$	
TOTAL TRANSIENT LOADS DURING ACCIDENT	σ_x	$\pm 36,700$	OUTER FIBERS; +TOP OF NOZZLE -BOTTOM
	σ_θ	$\pm 19,500$

FIGURE 2.32. *Stresses During Blowdown Transient
at Time of Accident*

The resulting stresses from this axial gradient alone, as well as coupled with a radial gradient, are given in Figure 2.33. The radial gradients were based on a flat slab approximation. The vessel has not yet been subjected to these thermal stresses since a top blowdown at 600 °F has not been made.

The second thermal condition, a situation considering water in contact with the nozzle for 45 sec, occurs for top blowdowns and some small nozzle bottom blowdowns. This condition, derived by imposing a thermal gradient on each thickness, is the same as that attained after 45 sec in a flat slab of identical thickness and thermal boundary conditions. The time of 45 sec was chosen by considering the time to maximum stress in a flat plate. The results are given in Figure 2.34. This condition, imposed upon the vessel in the past, is of some

MAXIMUM COMBINED BENDING AND MEMBRANE STRESSES,
NODES 5-17

STRESS	VALUE (psi)	LOCATION (NODE AND FIBER)		LOADING
σ_x	36,600	5	INNER	WITH RADIAL GRADIENT
σ_θ	39,200	5	INNER	WITH RADIAL GRADIENT
$2\tau_{\max}$	39,200	5	INNER	WITH RADIAL GRADIENT
TENSILE σ_x ON	27,000	11-15	INNER	WITH RADIAL GRADIENT

FIGURE 2.33. Stress Due to Temperature Gradients,
3.6 min. After Blowdown Ends

MAXIMUM COMBINED BENDING AND MEMBRANE
STRESSES, NODES 5-17

STRESS	VALUE (psi)	LOCATION (NODE AND FIBER)	
σ_x	41,780	5	INNER
σ_θ	41,950	5	INNER
$2\tau_{\max}$	41,950	5	INNER
TENSILE σ_x	39,350	11-15	INNER

FIGURE 2.34. Stresses Due to Temperature Gradients
45 sec After Blowdown Ends

importance as will be seen in the following discussion. It is also interesting to note that another method of stress calculation based on flat slab theory gives rise to much smaller tensile stresses on the inner surface than those predicted by SEAL SHELL for the spherical portion of the vessel (16,800 psi versus 42,000 psi). On the other hand, the compressive stresses in the outer fibers compare quite well (both about 8900 psi). This large difference is of little consequence except perhaps in the area where the fault plane emerges into the nozzle opening.

The third and final situation considered was an attempt to simulate the condition during the period when water remaining in the vessel after a bottom blowdown continues to boil until it is completely evaporated or until boiling ceases. Rough calculations indicate bulk temperature of the vessel bottom to approach that of the remaining fluid, i.e., about 200 °F within a period of 10 min or less. On the other hand, little heat is transferred across the nozzle surface since only steam is in contact there. Its temperature, therefore, is assumed to remain very close to the condition at the end of the blowdown and was assumed to be that prevailing at 40 sec after blowdown.⁽²⁾ This temperature profile ranges from 540 to 590 °F along the nozzle length. In short, the condition is one of a hot nozzle combined with a cool vessel bottom.

Because the actual nonaxisymmetric condition could not be analyzed with the tools at hand, the problem was solved by approximating a hot nozzle and a cool sphere. This approximation eliminates $\tau_{x\theta}$ and $\tau_{y\theta}$, but since $\tau_{x\theta}$ is not a critical stress component as discussed before, and since this approximation should result in higher than actual τ_{xy} stresses, the results can be considered as an upper bound of some value.

Two transition profiles were taken. One made the transition from the 200 °F temperature to the hot nozzle from nodes 4 to 14, and the other from nodes 9 to 14. These choices were made in an attempt to bracket the true condition. The results are given in Figure 2.35. This condition has occurred in the past and will occur in the future.

Failure Mode

With a weakened plane in the vessel wall and with the remaining material structurally sound, the failure mode most probably would result from propagation of the defect into and around the nozzle opening until the axisymmetric cross section appears as in Figure 2.36. Utilizing this geometry, the full pressure situation (2400 psi) was checked for the maximum stress values. The nominal membrane stress in the 1.5 in. thick portion of the shell is about 19,000 psi. The maximum combined bending-membrane stress from SEAL SHELL was found to be on the inner face at the junction of the 1.5 in. and 3.624 in. shells. This value was $2\tau_{\max} = 32,000$ psi. The maximum membrane stress, found to be about 14,500 psi or somewhat less than the 19,000 psi nominal, can be attributed to the "short span" involved and its ability to carry some of the load in shear. Since the 32,000 psi is probably greater than yield at 600 °F for SA 212 B (nominal yield is 28,100 psi but perhaps higher for this particular specimen), a redistribution of the stresses and an upward adjustment of the membrane stress toward the 19,000 psi value will occur.

Summary and Conclusions

The maximum stress conditions across the failure surface were found to be those associated with the hot nozzle-cool shell situation (maximum $\tau_{xy} = 8700$ psi) and the accident transients (maximum $\tau_{xy} = 8600$ psi). However, neither of these stresses reaches the yield point (14,000 psi, pure shear)

MAXIMUM COMBINED BENDING AND MEMBRANE STRESSES, NODES 5-17			
STRESS	VALUE (psi)	LOCATION	LOADING
σ_x	25,500	NODE 14 OUTER FIBERS	TRANSITION AT NODE 4
σ_θ	-42,450	NODE 14 INNER FIBERS	TRANSITION AT NODE 9
$2\tau_{\max}$	42,450	NODE 14 INNER FIBERS	TRANSITION AT NODE 9
TENSILE σ_x	ALL COMPRESSIVE	NODES 11-15	INNER

FIGURE 2.35. *Stresses in Nozzle C Due to Hot Nozzle - Cool Vessel Condition Approximates Conditions After a Bottom Blowdown at a Temperature of 600 °F*

and certainly do not come close to the ultimate strength (36,500 psi, pure shear). None of the other stresses, except the maximum bending stresses found in connection with the accident loads were considered significant. The value for σ_x (36,700 psi), by exceeding the tensile yield stress of 28,000 psi, leads to intensification of the mid-plane shear stresses. It is questionable whether the bending stresses are as high as indicated because bending results derived by Reference (3) methods for an axial load are indicated to be substantially over-estimated when compared to the SEAL SHELL

FIGURE 2.36. *Assumed Failure Mode of Nozzle C, Axial Cross Section*

method for handling axial load cases. On the other hand, the membrane stresses are higher in the SEAL SHELL results. This is not surprising since the shell treated herein falls out of the applicability range for Reference (3) results. However, even though these values are assumed to be correct, it still appears doubtful that shear stress τ_{xy} for the accident case would exceed yield, and it certainly would not exceed the ultimate. One other comment appropriate here is to note that, although the thermal bending stresses (45 sec case, Figure 2.34) are quite high, they are still within the code allowable.

The question of whether this defect propagation will continue is still to be answered. The shear value found in the hot nozzle-cool sphere case may, with some intensification, be sufficiently high to induce further defect growth. This possibility would be increased should a weakened plane throughout the vessel shell in the nozzle region be present. In fact, results previously described herein point strongly to the significant conclusion that, even for substantially underestimated loads, the failure would not have occurred had the vessel material been sound.

Consider again the assumed failure mode. "Turning" of the failure plane into the outer or inner surfaces, while possible, appears unlikely in the light of the stress intensities disclosed. It should also be noted that this possibility, while relating to crack propagation, was not subjected in this study to an "in depth" analysis. However, considerable yielding in SA 2128, generally considered a ductile material, should occur before tearing or cracking.

A crude limit analysis type of approach to the problem indicates that, based on an ultimate strength of 73,000 psi (conservative for this material) the 2400 psi internal pressure is only 19,000/73,000, or 0.26% of the ultimate failure pressure. Even with the higher bending stress ratio (32,000 psi at the 2400 psi pressure) maintained into the yielding region, this factor is still only 0.44. One final question is the possibility of a failure growth as extensive as that previously assumed. Two factors tend to discredit this possibility. The first is that little difference exists in stresses over, and those under, the nozzle. The most glaring exception here is the accident case where the change in sign of σ_y could have accounted for the difference in over or under behavior. Since no defects show up on the top side of the nozzle, this area can most likely be considered sound. Secondly, the 45 sec

thermal condition imposes a large (40,000 psi) tensile stress upon the area where the weakened plane would emerge into the nozzle opening. It would appear that the nozzle, having been previously subjected to this condition, should have failed because of the presence of this weakened plane and that a postaccident examination of the vessel should have detected it.

Many unknowns still remain and certainly there are many limitations to the analyses presented herein. However, assuming that the vessel material is sound and that the plane, although weakened, is reasonably ductile, it may be concluded that properly monitored testing of the defect can proceed with the degree of safety normally associated with high pressure vessel research.

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Homogeneous Blowdown Prediction - W. C. Townsend

A method of predicting the critical mass velocity, pressure, and quality at the exit of the blowdown nozzle as a function of the vessel pressure and entropy has been developed. This method is based on a homogeneous fluid and uses an approximate equation to represent an isentropic expansion process for water.* The method assumes an isentropic path and critical flow

* "Studies of Reactor Containment," IITRI-T-6042-25. October through November, 1966.

in the discharge duct. The technique, particularly useful in determining the flow rate by hand calculations will not, however, predict a vessel pressure-time relationship nor define the expansion process (i.e., isentropic etc.) in the vessel. The method predicts a critical mass velocity from the vessel pressure identical to that obtained from a homogeneous equilibrium critical flow model.

The approximate equation used to represent the isentropic expansion is given by:

$$\left(\frac{1}{P_1} - \frac{1}{P_2} \right) U_o^2 = v_1 - v_2 \quad (1)$$

where P_i : pressure at plane i

v_i : specific volume at plane i

U_o^2 : proportionality factor

This relation was found to hold in the pressure range of interest (2500 psig to 500 psig). The proportionality factor, U_o^2 , is dependent only on the entropy of the fluid in the nozzle. Equations of the expansion path and energy were substituted into the definition of critical mass velocity

$$\left(\frac{\partial G}{\partial P} \right)_s = 0 \quad (2)$$

to result in an equation

$$\left[1 - \frac{P^*}{P_v} \left(1 - \frac{C}{U_o} \right) \right]^2 = -2 \left[\left(1 - \frac{C}{U_o} \right) \left(1 - \frac{P^*}{P_v} \right) + \ln \frac{P^*}{P_v} \right] \quad (3)$$

relating the term C/U_o to the critical pressure ratio, P^*/P_v , where P^* is the critical pressure at the exit and P_v is the vessel pressure. The term C/U_o (C is the local acoustic velocity) is dependent only on the fluid and is given by

$$\frac{C}{U_o} = \frac{P_v}{U_o^2} \quad (4)$$

Then, given the fluid state in the vessel, a C/U_o term may be calculated which will, in turn, give a critical pressure ratio from Equation (3). The velocity and specific volume and therefore, the mass velocity at the critical state may then be calculated from the critical pressure and upstream conditions.

This method has been used to calculate the relationship between the critical mass velocity and vessel pressure for two different expansion processes occurring in the vessel. The results are shown in Figure 2.37. The first case, given by the solid line, is where the vessel fluid follows the saturated liquid curve during decompression. The second case, given by the dashed lines is where the vessel fluid expands isentropically from a given saturated liquid state. Two specific saturation states (saturated liquid at 1090 psig and at 1540 psig) have been considered and are shown in Figure 2.37 as dashed lines.

Comparison of results from this method with CSE experimental data from two blowdowns having nominal 2 in. (Run B-10) and 4 in. (Run B-12B) diameter breaks, is also shown in Figure 2.37. Each of these comparisons was based on the vessel pressure and entropy at the time that the vessel fluid reached saturation. This pressure is defined by the intersection of experimental pressure and temperature data with the saturation curve. The experimental data from Run B-10 (2 in. break diameter) approximate the curve on which the vessel fluid is following the saturated liquid curve during decompression. Data from Run B-12B (4 in. nominal break diameter) agree with the condition that the vessel fluid expands isentropically. The apparent oscillation noted in the experimental data of Run B-12B is believed to result from the method of reducing the mass velocity data.

FIGURE 2.37. Mass Velocity Prediction

FIGURE 2.38. Nozzle Pressure Prediction

The vessel fluid expansion process is noted to differ between Runs B-10 and B-12B. The entrance fluid for Run B-10 seems to be a saturated liquid, while the entrance fluid for Run B-12B, a two phase fluid with a quality greater than zero, implies the entrainment of vapor and emphasizes the need to define quality of the fluid entering the discharge duct.

The predicted critical pressure is compared in Figure 2.38 with experimental nozzle pressure data from Run B-12B. The predicted critical pressure is within 10% of that obtained experimentally except at pressures less than 200 psig above the vessel pressure at transition from two-phase fluid to vapor flow.

Supporting Analytical Studies - W. A. Carbiener

Battelle-Columbus is performing analytical studies in support of the CSE blowdown program. The initial task is to evaluate the relative effects of parameters including coolant quality gradient in the core, heat output from the core, and external coolant loop volume on the pressure differentials to which the reactor vessel intervals may be subject during coolant blowdown.

A single node quasi-steady state computer model was written to perform these preliminary studies. This model, relatively simple in comparison to the larger, more complicated models (i.e. FLASH, BURST, etc.), affords a convenient tool for performing initial parametric investigations. The basic model has been modified to include two nodes, phase separation of the fluid, specification of break locations, and subcooled and superheated fluid properties.

These models have been used to study the effects of coolant quality and quality gradient on the initial critical flow, momentum, and velocity head across a sieve plate

inserted in the simulator vessel. The initial quality gradient condition, resulting in larger forces on the sieve plate than does a constant quality case, is related to quality effects on critical mass flow rates and on acoustic velocity.

FISSION PRODUCT AEROSOL CONTROL - L. C. Schwendiman and
H. H. Van Tuyl

Removal of Organic Iodide with Reactive Sprays -
L. C. Schwendiman and A. K. Postma

Volatile and gaseous fission products will be released to the containment volume following a serious reactor accident. A possible method for reducing the airborne concentration of radioiodine and its compounds is to immobilize them by adding a reactive compound such as hydrazine to the spray.

During the current reporting period, the spray washout of methyl iodide has been measured at temperatures up to 123 °C, experimental measurements have been compared to theoretical predictions, and a method was developed for more accurately assessing drop absorption.

Three spray washout experiments were carried out at temperatures near 120 °C to demonstrate the effect of higher temperature on methyl iodide absorption rate, and on the reaction of hydrazine with oxygen. These runs were conducted in a pressure vessel 3 ft in diameter and 8 ft tall (PAT). The composition corresponded to a saturated air-steam mixture with an air partial pressure of one atmosphere at 20 °C.

Drops were generated from an array of 9 impingement nozzles. Drop size produced by this nozzle system was measured at room temperature and atmospheric pressure by an oil collection method. The mass median diameter of the drops at a point 8 ft below the nozzles was measured to be 340 μ . This size, more than twice the diameter for droplets from a single nozzle of the

same type, indicated drop coalescence to be appreciable. The effective drop size would thus be smaller than the size indicated by the measurements.

The results of the high temperature spray runs are summarized in Table 2.II.

TABLE 2.II. *Summary of High Temperature Washout of CH_3I by Hydrazine Sprays*

Run No.	Temperature, °C	Spray Density, gpm/ft ²	Initial Spray Solution Composition	Measured Washout Half-Time, min
HT-1	120	0.08	0.06 M NH_4OH 10 wt% N_2H_4 3000 ppm H_3BO_3	8
HT-2	120	0.12	0.06 M NH_4OH 6.5 wt% N_2H_4 3000 ppm H_3BO_3	9
HT-3	123	0.085	0.05 M $NaOH$ 10 wt% N_2H_4 3000 ppm H_3BO_3	7

The spray densities shown in Table 2.II were calculated on the basis of the total spray rate. With roughly 30% of the sprayed liquid reaching the wall as a film, the drop fall rate would be about 70% of the values shown in Table 2.II. The washout rate measured is the result of absorption by both the falling drops and the wetted wall. The relative absorption by drops and wall film was assessed in Runs HT-2 and HT-3 by separately collecting the liquid drained down the wall and that collected in the bottom of the tank. Analysis of these two liquid volumes indicated that roughly 50% of the absorption occurred in the wall film. On a volume basis, the wall liquid was 2 to 3.5 times as effective as the falling drops.

The washout half-times for the experiment reported here correspond to washout rates about 5 times faster than those measured in earlier runs at atmospheric pressure and at temperatures near 90°C. Thus, the positive temperature coefficient of the spray washout rate is probably due primarily to the rapid increase in solution reaction rate with temperature.

The presence of borate ion, a potential additive to spray solutions, is not indicated by the measured washout rates to adversely affect the removal of methyl iodide by hydrazine sprays. Previous spray experiments carried out at lower temperatures had shown no appreciable effect by borate ion upon the washout rate. In previous runs, ammonium hydroxide had been added to the spray solution to maintain it basic. In Run HT-3, however, sodium hydroxide was used with negligible influence on the overall washout rate. Reaction kinetic studies have indicated the rate constant for the solution reaction to be largely independent of pH. Hence, it is anticipated that the spray removal rate would be independent of basicity of the spray solution.

Hydrazine concentration in the spray solutions was measured as a function of time, and chemical reaction between hydrazine and oxygen was concluded to be relatively slow. In Run HT-3, for example, less than 10% of the hydrazine was lost from the liquid system during a 1 hr recirculation spray period.

Spray washout rates were predicted from absorption theory for a pseudo first order liquid phase reaction. The absorption rate for the wall film, calculated on the basis of the penetration theory, and drop absorption, calculated from the stagnant drop model, allowed for first order chemical reaction. Because solubility and reaction rate data have not yet been obtained at the temperatures used in the spray run, extrapolation of data has been required. Using extrapolated values of

solubility and reaction rate, the calculated washout half-times are 4 to 5 times longer than the measured values. Extrapolations as great as those required here are prone to inaccuracies and might therefore account for the discrepancy between measured and predicted washout rates. Thus, at present, the comparison with theory cannot be considered satisfactory until solubility and reaction rate measurements at higher temperature are obtained.

A study of drop absorption is presently in progress to provide direct measurement of absorption by falling drops. To date, methods of assessing wall versus drop absorption have embodied the separate collection of the wall film and the drops. This technique is open to question because the drops spread as a film at the bottom of the spray chamber where additional film absorption will occur.

A drop collector has been designed to circumvent this problem. This collector will be placed inside the spray chamber, and drops falling into a flask with a vertical chimney will be separated from continued contact with airborne methyl iodide by a purge of clean air. Use of this drop collection method, expected to provide direct measurement of the drop absorption per volume per unit gas phase concentration, is directly related to the spray washout rate.

Physical Chemistry of Methyl Iodide Systems - R. A. Hasty and S. L. Sutter

The objective of this research is the determination of physiochemical factors affecting the spray removal rate of methyl iodide in reactor containment atmospheres. Earlier studies have demonstrated the complex relationship of the removal rate from the gas phase to depend upon properties of the reactive solution used as the spray.⁽¹⁾ A model involving

the dissolution of methyl iodide at the gas-liquid interface, diffusion into the droplet of spray, and reaction within the droplet has been proposed to mathematically depict the case of spray removal of methyl iodide. The partition coefficient of methyl iodide in the concentration range of interest, i.e., 10^{-7} to 10^{-8} moles/liter (1.4 to 14 ppm), was also measured from 5 to 68 °C.⁽²⁾ To provide data necessary for evaluation of the spray removal model, the rate of reaction in solution of methyl iodide is needed, particularly at elevated temperatures. Measurements at room temperature and an estimate of the rate at 50 °C were reported earlier.

The sparge removal technique, described briefly in the preceding report, was utilized to determine the rate of reaction of methyl iodide in aqueous solution.⁽³⁾ The concentration of unreacted methyl iodide is determined from this technique by the concentration of methyl iodide in the sparge gas leaving the reaction vessel. Thus, the high volatility of methyl iodide which has presented so much of a problem in the studies of its behavior in aqueous solution (because of the unfavorable partition coefficient) is utilized to "observe" the reactions. The rate of disappearance of methyl iodide is given by two terms, both of which are first order with respect to the methyl iodide concentration

$$-\frac{dc}{dt} = k_r C + k_s C \quad (1)$$

where k_r is the pseudo first order reaction rate constant for the reaction, k_s is the sparge removal rate constant, and C is the concentration of methyl iodide in solution.

Since the gas phase concentration of methyl iodide is proportional to the solution concentration, we can substitute into Equation (1) the gas phase concentration, integrate, set the boundary condition (initially C_g equals C_g^0), and obtain

$$A_s = \frac{[dv/dt] C_g^0}{(k_r + k_g)} \quad (2)$$

where $[dv/dt]$ is the flow rate of sparge gas and A_s is the amount of methyl iodide sparged from the reaction cell. Similar treatment for no reaction in the solution, i.e., with k_r equals to zero, yields the amount of methyl iodide introduced to the solution, A_o . Combination of these two expressions yields the relationship used to calculate the reaction rate constant.*

$$\frac{1}{(k_r + k_s)} = \frac{1}{k_r} - \frac{1}{k_r} \frac{A_s}{A_o} \quad (3)$$

The observed rate of decrease of methyl iodide, $(k_r + k_s)$, and the amount of unreacted methyl iodide recovered, A_s , are determined in a series of experiments where the flow rate of sparge gas is varied. The rate of methyl iodide reaction in solution is obtained from the intercept of a data plot in the format given by Equation (3).

The pseudo first order rate constants thus obtained in a 0.053N sodium hydroxide solution (0.2 wt%) at 34.7 °C and $\sim 3 \times 10^{-8}$ moles/liter methyl iodide (0.5 ppm) for 0.54, 1.19, 2.4, and 4.8 wt% hydrazine solutions are 0.00063, 0.00123, 0.0027, and 0.0053 sec⁻¹. The second order rate constant is therefore $(3.5 \pm 0.2) \times 10^{-3}$ liters mole⁻¹ sec⁻¹ at 34.7 °C (i.e., 0.0035 sec⁻¹ for a 3.2 wt% hydrazine solution).

Similarly, the second order rate constant for the reaction of hydrazine and methyl iodide at 24.6, 49.3, and 64.7 °C are 0.001, 0.015, and 0.064 liter mole⁻¹ sec⁻¹, respectively. The activation energy for the reaction is 20.2 ± 0.4 kcal/mole.

* The technique will be described in detail in the next topical report on this work.

At both 49.3 and 64.7 °C, the reaction was complicated by the hydrolysis of the methyl iodide by the solvent. This contribution, the hydrolysis by solvent, becomes important only when very dilute concentrations of hydrazine are used. The hydrolysis at 49.3 °C contributes ~20% for a 0.01 mole/liter hydrazine solution (0.03 wt%) and only 2% for a 0.1 moles/liter hydrazine solution (0.3 wt%). The magnitude of this hydrolysis compares very closely to that calculated from the value reported in the literature.⁽⁴⁾

The hydroxide ion concentration had little effect on the rate of reaction, although a decrease of 8% was observed going from a sodium hydroxide concentration of 0.05 moles/liter to a hydrogen ion concentration of 0.26 moles/liter at 34.7 °C.

Similar studies on the effect of carbonate and borate ions on the reaction are underway. Also, the rate of reaction of substituted hydrazine are being examined.

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3. PRESSURE BEARING COMPONENT EVALUATION AND MONITORING STUDIES

CRACK DETECTION IN PRESSURE PIPING BY ACOUSTIC EMISSION -

P. H. Hutton, R. N. Ord, H. N. Pedersen, and J. C. Spanner

Introduction

Enhancement of public safety through development of improved techniques for integrity surveillance of nuclear reactor pressure boundaries is an area of considerable merit. Detection of acoustic emission or elastic waves produced in metal as it deforms and fractures is the basis for such a development program at the Battelle Memorial Institute/Pacific Northwest Laboratory. This technique would provide a warning of incipient failure in time to take necessary remedial action.

The program began in February, 1966 under the sponsorship of the USAEC's Division of Reactor Development and Technology. A coordinated effort evolved between this program and acoustic emission work being done by Phillips Petroleum Company is aimed at increased overall efficiency in developing a tested integrity surveillance system utilizing acoustic emission.

The program effort for this quarter, as in previous reports is discussed under six task headings. Low alloy carbon steel was the primary test material. However, some work also has been done with Type 304L SS.

Discussion

Task 1 - Acoustic Emission Signal Analysis and Characterization

Characterization of acoustic emission propagation behavior in various media is being studied. This work has demonstrated the manner in which reflected signals enter into a single

emission pulse. A simulated acoustic emission pulse is shown in Figure 3.1 with its characteristic fast rise response, and with a slow decay in amplitude lasting several milliseconds. This pulse was obtained by transmitting a 0.5 μ sec square wave pulse through a PZT thickness expansion mode crystal mounted perpendicular to the face of a 0.25 in. thick 304L SS plate. A receiving crystal with similar characteristics was located at selected distances from the transmitting sensor along the surface of the plate. Expansion of the pulse, as shown in Figure 3.2, reveals a complicated wave form modulating the carrier frequency. The major envelope peaks have been correlated with preferred paths of acoustical wave propagation within the specimen geometry. These transmission paths are shown in Figure 3.3 and Figure 3.4 for two different receiving sensor locations. The envelope peaks correlate with a velocity equal to 1.155×10^5 in./sec, approximately equal to the Rayleigh mode of propagation. Each envelope is 20 to 40 μ sec in duration, with the envelope representing a distribution of mode converting waves propagating down the thickness of the plate. The duration and velocity at which the envelope can be detected is subject to the mounting direction angle of the transmitting and receiving sensor. Fourier analysis theory shows the width of the spectral band to be inversely proportional to the pulse duration which, for this test, was 1 MHz. Spectral components above 200 kHz were attenuated by the strong resonant characteristics of the transmitting and receiving crystals.

Because of the influence of the transmitting sensor on the spectral content and directionality of the transmitted wave front, a test suitable as a controlled source of emission duplicating a flaw growth mechanism in metal was sought. A test was performed by using a pulsed laser focused into a Lucite block with sensors mounted in the appropriate directions to measure

2 msec/cm

EXPANSION OF ACOUSTIC EMISSION PULSE

TX } 240 Mil
RC } 5 Meg PZT-5 Crystals

.05 V/cm

12" Apart

.2" Aluminum Plate

1 msec/cm

.1 msec/cm

FIGURE 3.1. Expansion of Simulated Acoustic Emission Pulse

FIGURE 3.2. Path Propagation from Acoustic Emission Pulse

$$V = 1.155 \times 10^5 \text{ in/sec.}$$

$$T(1) = \frac{27}{V} = 234 \mu\text{sec.}$$

$$T(2) = \frac{27+8+8}{V} = 372 \mu\text{sec.}$$

$$T(3) = \frac{11+11+27}{V} = 424 \mu\text{sec.}$$

$$T(4) = \frac{26.5+26.5}{V} = 458 \mu\text{sec.}$$

$$T(5) = \frac{31+20+11.5}{V} = 541 \mu\text{sec.}$$

$$T(6) = \frac{11+11+27+8+8}{V} = 562 \mu\text{sec.}$$

$$T(7) = \frac{14.5+19+33}{V} = 576 \mu\text{sec.}$$

$$T(8) = \frac{13.5+27+28.5+10}{V} = 670 \mu\text{sec.}$$

$$T(9) = \frac{24+47.8+24}{V} = 823 \mu\text{sec.}$$

FIGURE 3.3. Acoustic Emission Propagation Paths in a Test Plate (1/4 in. thick, 304L SS)

$$V = 1.155 \times 10^5 \text{ in/sec.}$$

$$T(4) = \frac{15.5+16.5+32.5}{V} = 558 \mu\text{sec.}$$

$$T(1) = \frac{24}{1.155 \times 10^5 \text{ in/sec}} = 208 \mu\text{sec.}$$

$$T(5) = \frac{11+11+24+11+11}{V} = 589 \mu\text{sec.}$$

$$T(2) = \frac{11+11+24}{V} = 398 \mu\text{sec.}$$

$$T(6) = \frac{13.4+27+28.5+12.8}{V} = 706 \mu\text{sec.}$$

$$T(3) = \frac{26+26}{V} = 450 \mu\text{sec.}$$

$$T(7) = \frac{24+47+24}{V} = 820 \mu\text{sec.}$$

FIGURE 3.4. Acoustic Emission Propagation Paths in a Test Plate (1/4 in. thick, 304L SS)

longitudinal and shear components of the photon induced emission. Figure 3.5, a photograph of the block showing the photon induced flaws, is considered a reasonably good representation of a cleavage failure in metal from the standpoint of the signal generated - i.e., an internally generated shock signal. A resulting longitudinal wave detected at the specimen surface as recorded on an oscilloscope directly and on playback from a tape recorder is shown in Figure 3.6. This comparison demonstrates the distortion incurred in a high frequency signal by the limitations (300 kHz) of the tape recorder. The initial spike, an electrical transient produced by the discharge of the Laser, occurred approximately when the flaw was formed in the Lucite block. The acoustic emission wave front occurred about 26 μ sec later. Of major significance is the fact that the direct recording shows an initial high spectral content up to 1 MHz on the leading edge, followed by lower frequencies produced by the specimen geometry and sensor resonant conditions. This leading edge information appears to be controlled by the source mechanism and to be independent of specimen geometry. As discussed later, similar signal characteristics were observed in ductile pipe rupture tests. These were of a lower frequency due to the ductile failure mechanism.

Initial work on selected mode detection through the use of critical angle orientation of the transducer shows considerable promise. The major benefit of this technique, serving to enhance the accuracy of source location by triangulation, is accomplished by the fact that all detected signals are of one mode and, hence, of a constant propagation velocity. A sharper wave front is another benefit of this technique, application of which is dependent on incorporating omnidirectional capability through the use of a transducer adapter or specially shaped transducer element.

FIGURE 3.5. *Lucite Block Test Arrangement Used to Simulate
Acoustic Emission Pulses*

Direct Record

Through Ampex Tape Recorder

5 V/cm
 Top 200 μ sec/cm
 Bottom 20 μ sec/cm
 CHAN 4
 $x = 0$ mm
 Pulse 25

FIGURE 3.6. *Laser Induced Emission Pulses from Lucite Block*

The critical angle technique has been used to study acoustic emission from small carbon steel and austenitic stainless steel samples. In this preliminary work, some frequency components as high as 10 MHz were detected.

Task 2 - High Temperature Transducer Development

The PZT-5A transducer assembly in use for acoustic emission study work has been analyzed in terms of basic components to improve the sensitivity and response bandwidth. Figure 3.7 shows the comparative response of a 0.240 in. diam, 1 MHz, PZT-5A crystal mounted (1) bare, (2) encased in a brass housing with epoxy backing, and (3) encased in a brass housing with epoxy backing and an epoxy front lens. It is apparent that the bare crystal has a prominent radial resonance at 370 MHz (the resonance at 230 kHz is a characteristic of the transmitting crystal used.) Addition of the epoxy backing reduces the resonant response "Q" from about 200 to 20 (or by 90%), while the nonresonant response is reduced by only about 20%. The front lens, in addition to the epoxy backing, while producing only a slight additional damping of the resonant response, reduces the nonresonant response by about 80%. These comparisons lead to the conclusion that the encased crystal with backing and without a front lens is the best combination for broadband study work.

A further step in the study of sensor characteristics was to compare the spectral content of a 0.125 and a 0.240 in. diam PZT-5 expander mode crystal mounted with RTV-116 silicone rubber couplant on a plate. The sensors were mounted perpendicular to the plate at a separation of 24 in. The spectral content between 0 and 1 MHz, and location of the transmitter and receiver in each case, is illustrated in Figure 3.8. These tests, also run with the sensors interchanged, showed no significant difference in results. Study of Figure 3.8 shows the

FIGURE 3.7. Sensor Fabrication Study

Frequency Meg HZ
 .5 mV/cm
 TX West 125 Mill, 1 Meg Crystal
 RC East 240 Mill, 1 Meg Crystal

Frequency Meg HZ
 .5 mV/cm
 TX West 125 Mill, 1 Meg Crystal
 RC East 125 Mill, 1 Meg Crystal

.2 in. Aluminum
 Plate
 RTV Bond
 24 in. Separation

Frequency Meg HZ
 .5 mV/cm
 TX West 240 Mill, 1 Meg Crystal
 RC East 125 Mill, 1 Meg Crystal

Frequency Meg HZ
 2 mV/cm
 TX West 240 Mill, 1 Meg Crystal
 RC East 240 Mill, Meg Crystal

FIGURE 3.8. *Spectrum Analysis of PZT Crystal Diameter Mode Study*

difference in characteristics between the 0.125 and 0.240 in. crystal. Reference to the radial disc mode of vibration for each crystal, a value of 350 kHz, is obtained from the 0.240 in. diam crystal and 570 kHz for the 0.125 in. diam crystal. It is suspected that the 220 kHz resonance is a cross coupling mode in the 0.125 in. crystal. The 0.240 in. diam crystal gives the greatest sensitivity in this thickness (1 MHz) crystal, and this comparison demonstrates no adverse spectral characteristics compared to the 0.125 in. diam crystal. On this basis, the 0.240 in. diam crystal is being used in sensor fabrication.

A final step in this study was to evaluate the effect of various thicknesses of RTV-116 silicone rubber coupling material on the received signal characteristics. Figure 3.9 demonstrates the reduction of received signal incurred between a thin coat less than 0.001 in. thick, a coat 0.065 in. thick, and a coat 0.750 in. thick. In all cases, the RTV was cured according to specifications supplied by the manufacturer. A reduction in signal sensitivity of about a factor of 3 is indicated between the 0.001 in. and the 0.065 in. bond, and a reduction of about a factor of 10 is indicated between the 0.001 in. and the 0.750 in. bond. Figure 3.10 indicates the effect of coupling material thickness on the spectral resonant peaks. The thicker layers show a significant amount of damping at the resonant points as well as a reduction in general sensitivity. The thin (0.001 in.) layer of silicone rubber currently in use for acoustic coupling appears to be the best choice.

Long term tests of PZT-5A transducer longevity in a 600 °F environment currently in progress have accumulated 500 hr of exposure to date with no apparent deterioration of the crystal. The epoxy backing material currently in use is

< 1 Mill RTV
 .05 V/cm
 50 μ sec/cm

0.5 in. - A 212 B Plate
 125 Mill, 1 Meg HZ
 TX and RC
 RTV Bonding

~ 65 Mill RTV
 .02 V/cm
 50 μ sec/cm

~750 Mill RTV
 .02 V/cm
 50 μ sec/cm

FIGURE 3.9. Bonding Thickness Sensor Study

Frequency~Meg HZ
 <1 Mill RTV
 .5 mV/cm

.5 in. -A-212-B Plate
 125 Mill, 1 Meg HZ
 TX and RC
 RTV Bonding

Frequency~Meg HZ
 ~65 Mill RTV
 .2 mV/cm

Frequency~Meg HZ
 ~750 Mill RTV
 .2 mV/cm

FIGURE 3.10. *Spectrum Analysis of Bonding Thickness Study*

not completely satisfactory for high temperature service due to the rather high coefficient of thermal expansion. A search is underway for a substitute material with similar bonding characteristics but with lower thermal expansion.

A transducer assembly using lithium niobate (LiNbO_3) piezoelectric material is being fabricated for service to at least 1000 °F. The sensitivity of the LiNbO_3 crystals has been enhanced to a point considered useable for acoustic emission detection. This material has the added merit of being much less prone to high "Q" internal resonances. The assembly will be tested for resistance to high temperature and nuclear radiation.

Task 3 - System Analysis Study

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

Task 4 - Monitor System Development

There were no significant developments under this task. Work was limited to overcoming a filter roll-off deficiency in the solid state signal conditioning system described in the last quarterly report. Some effort has also been directed toward developing a low noise, broadband preamplifier beyond the capabilities of the one presently in use. This objective is consistent with the recognized need to investigate acoustic emission in the low MHz range.

Task 5 - Cooperative Work with Phillips Petroleum

PNL and Phillips Petroleum Company personnel devoted a considerable amount of joint effort toward compiling answers to a series of questions concerning acoustic emission work and to preparing a joint program outline as requested by DRDT.

The completed package, transmitted to DRDT March 15, 1968, contained a complete program outline aimed at achieving three successive levels of refinement in acoustic emission monitor systems. These are:

- Detect and locate flaws during hydrostatic proof testing of either individual reactor components or the assembled reactor system.
- Detect and locate flaws on a continuous, on-line basis on an operating reactor pressure system.
- Detect, locate, and quantitatively measure flaws on a continuous, on-line basis on an operating reactor pressure system.

The general division of effort is based on research and development work being done by PNL, and plant application testing being done by Phillips.

Task 6 - Application Engineering

Two pipe burst tests (No. 8 and 9) performed by Battelle-Columbus in mid-December, 1967 as part of their program to investigate ductile pipe rupture, were monitored by PNL for acoustic emission. Both test specimens were fabricated of 24 in. OD Schedule 100, Type A-106-B carbon steel pipe with a part-through-wall milled flaw employed in both tests. Only the acoustic emission aspects of the tests are discussed here. The other test results are discussed in a BMI quarterly report.*

The system utilized to measure acoustic emission in both tests consisted of six PZT-5A, 5 MHz, shop made transducers mounted on the specimen using lavite holders attached to the surface with machine screws. Figure 3.11 shows transducer

* R. J. Eiber, et. al. "Investigation of the Initiation and Extent of Ductile Pipe Rupture," Quarterly Progress Report, October-December, 1967, BMI-1828. Battelle Memorial Institute, Cleveland, Ohio.

FIGURE 3.11. *Transducer Mounting Pattern Battelle-Columbus
Pipe Burst Tests*

location. The output of the transducers was connected to Ithaco preamplifiers with 25-ft long Microdot low noise cables. Preamplifier output was connected to a solid state six-channel, shop made signal conditioner through 300-ft lengths of Beldon coaxial cables. The signal conditioner provided an adjustable 24 dB/octave high-pass filter and another amplification stage. All six channels of information were then recorded on an Ampex FR-1300 magnetic tape recorder. Overall system gain was 1×10^4 and the filters were operated at 25 kHz. One channel of data was also routed to an emission rate analyzer-recorder where the emission rate was recorded on a strip chart. The emission rate recorder was operated continuously throughout the test. The tape recorder, however, was operated only intermittently until the emission rate began to increase about 8 min prior to failure at which time the tape recorder was started and operated continuously until the specimen failed.

A large number of very low level signals attributed to metal deformation were observed in both tests. The abundance of these signals relative to the test sequence followed a very logical pattern and were a major factor in determining the shape of the emission rate curves shown in Figures 3.12 and 3.13. The preliminary peak in these curves occurred about 4 min prior to failure in both tests. (NOTE: Failure pressure for Test No. 9 was erroneously stated as 1405 psig in the previously referenced BMI quarterly report. The correct figure is 1360 psig.) Typical examples of these signals are shown in Figure 3.14. The significant aspect of this part of the data is the fact that it controlled the shape of the emission rate curve. The resultant shape was very similar to that obtained when a tensile specimen is pulled. A tensile specimen produces a curve showing an increasing emission rate up to the onset of plastic deformation at which point the rate

FIGURE 3.12. Acoustic Emission Rate Versus Vessel Pressure-Final
9 min of Burst Test

FIGURE 3.13. Acoustic Emission Rate Versus Vessel Pressure-Final
 7 min of Burst Test

Typical Ductile Fracture Signal

3 1/2 Volts @ ≈ 120 kHz

0.0005 sec/cm Sweep Rate

0.2 Volts @ 25 kHz

0.0002 sec/cm Sweep Rate

0.4 Volts @ 25 kHz

0.0002 sec/cm Sweep Rate

Typical Deformation Signals

FIGURE 3.14. Example of Deformation and Fracture Signals Observed in Pipe Rupture Tests at Battelle-Columbus

tends to level out. As the specimen starts to neck and deformation becomes localized, the overall emission rate drops until the moment of fracture at which point the emission rate, of course, rises very abruptly. It would thus appear that the acoustic emission monitor in these burst tests was able to detect the sequence as the metal in the area of the flaw was loaded to yield, to the early stages of plastic deformation, to localized deformation and, finally, to failure. These results can be contrasted with the emission rate pattern derived from an earlier, similar test where a through wall flaw was used (Test No. 5). With this type of flaw, the amount of material undergoing plastic deformation prior to crack growth would be expected to be less than for a part-through-wall flaw. This conclusion was reflected in the emission rate curve for Test No. 5, controlled by burst signals from progressive crack movement. A general increase was observed through the last 10 min of the test as the crack grew to critical length and failure occurred. The lower level signals, indicative of metal deformation, were not evident in this earlier test.

Emission rate response to deformation signals during Tests No. 8 and 9 demonstrates the potential of using this information for early warning of flaw development under appropriate condition. These signals show a sharp rise front followed by a few cycles of low frequency (≈ 25 kHz) oscillation which may make them susceptible to masking by a noisy background. Their use in surveillance of a reactor system thus appears to be limited to hydrostatic test conditions.

A reasonable amount of effort to triangulate to a source location using these signals proved fruitless for the reason that the detected level of these signals was too low to enable a given signal to not appear consistently on multiple channels of the recorded data. An increase in transducer

sensitivity by about a factor of five since these tests were conducted should alleviate this problem by significantly improving the signal-to-noise ratio.

Evidence of a small amount of slow crack growth was detected in both specimens prior to failure. The location and timing of these indications is tabulated in Figure 3.15.

The burst signals permitting triangulation to the flaw as a source and indicating flaw extension exhibited a unique characteristic. Each of these bursts contained a packet of high frequency (≈ 100 kHz) energy at the leading edge. This packet persisted for only about 0.1 msec and then the signal converted to a low (≈ 25 kHz) frequency lasting for several milliseconds. In Figure 3.16, a typical example of these signals is contrasted with an extraneous burst present in the gross data. This leading edge packet is not simply the early signal arrival by the longitudinal mode because the transmission velocity is too low. Consideration of these data, together with recent laboratory tests to study wave propagation from impulse sources, indicates the leading edge packet to be the only portion of the burst signal indicative of the signal source. The rest of the signal is a function of specimen geometry. The thickness resonance of the specimen, in this case calculated to be 25 to 30 kHz, is borne out by the fact that the trailing part of the burst signals, the deformation signals, and all of the extraneous signal exhibit such a frequency.

Samples of the data trace at various points during the final part of Tests 8 and 9 are shown in Figures 3.17 and 3.18. Study of the information from Burst Tests 8 and 9 pointed out several areas of the acoustic emission monitoring technique requiring further development. The improvement of transducer sensitivity to reliably utilize deformation emission will enhance the signal-to-noise ratio and thus allow better

1. 2.4 Min. Prior to Failure, 2712 psig
2. 2.0 Sec. Prior to Failure, 2796 psig
3. Initiation of Failure, 2800 psig

1. 0.8 Min. Prior to Failure, 1319 psig
2. 0.7 Min. Prior to Failure, 1323 psig
3. Initiation of Failure, 1360 psig

FIGURE 3.15. Location of Flaw Increments Indicated by Acoustic Emission During Pipe Burst Tests at Battelle-Columbus

SENSOR NO. 4
2 VOLTS/cm, 0.1 mil-sec/cm
25 kHz HI-PASS FILTER
1323 psig (SEE FIGURE 13)
ACOUSTIC EMISSION SIGNAL

SENSOR NO. 4
2 VOLTS/cm, 0.1 mil-sec/cm
25 kHz HI-PASS FILTER
1355 psig (SEE FIGURE 13)
EXTRANEEOUS SIGNAL

FIGURE 3.16. *Comparison of an Acoustic Emission Signal from Flaw Growth and an Extraneous Signal Observed During Battelle-Columbus Pipe Burst Test No. 9*

detection of low level signals. As mentioned earlier, this improvement has already been accomplished to the extent of a factor of five. A problem in this improvement is introduced, however, by the stronger burst signals saturating the electronic system. A log suppression device to overcome this problem by attenuating the higher amplitude signals is being developed.

It was also observed that a transducer located just over the corner of a hemispherical end cap will see a different wave propagation mode and, hence, a different velocity than a transducer located on the body of the vessel. As used in these tests, the transducers on the body of the vessel were detecting Rayleigh waves while the one on the head was sensing shear waves. Wave propagation studies in progress are directed toward the selection or favoring of a particular propagation mode for detection at all points.

In summary, these tests are considered to contribute much information useful to the development of acoustic emission monitoring techniques. The use of a part-through-wall flaw presents

2520 psig

2710 psig

30 - 200 kHz B. P. Filter Scope Settings
0.2 V/cm, 2.0 msec/cm
Refer to Figure 12

2793 psig

2800 psig

FIGURE 3.17. Data Trace Samples from BMI Burst Test No. 8

1170 psig

1240 psig

30 - 200 kHz B. P. Filter Scope Settings
0.2 V/cm, 2.0 msec/cm
Refer to Figure 13

1290 psig

1360 psig

FIGURE 3.18. Data Trace Samples from BMI Burst Test No. 9

a condition more analogous to conditions possibly arising in field application to a pressure system than would a specimen with a through-wall flaw. The failure sequence with the part-through-wall flaw is more subtle from the acoustic emission standpoint. However, the acoustic emission system as used was still able to show distinct phases of the process even under the adverse conditions of boiling water noise and remote monitoring (300 ft lead wires). The system improvement work in progress, it is felt, will substantially improve monitoring effectiveness.

Installation of instrumentation on Commonwealth Edison's Dresden I nuclear power reactor for examining the background noise spectrum during operation has been arranged. Much needed information on the noise competition expected in monitoring acoustic emission from one of the major power reactor types (boiling water) during operation will thus be provided. Measurements will be taken during reactor start-up and during full power operation.

Work Planned for the Next Quarter

- Perform noise measurements on the Dresden I Reactor.
- Continue transducer development work.
- Monitor acoustic emission produced during one pipe fatigue test at General Electric Company, Vallecitos plant.
- Initiate a series of tests to support frequency classification of acoustic emission.
- Perform tests at NRTS, Idaho, to give a direct comparison of response characteristics of the PNL and the PPCo. acoustic emission monitor systems.

4. ENVIRONMENTAL STUDIES

DISPOSAL OF REACTOR OFF-GAS INTO SOIL SYSTEMS - L. G. King

Efforts have been directed during the past two years toward a field demonstration of the concept for disposal of reactor off-gas into the ground. This work has included planning, conducting, and analyzing a tracer test at a field site within the National Reactor Testing Station (NRTS) in Idaho. Battelle-Northwest's contribution to the overall project has been the mathematical modeling of gas flow during the injection, in providing advice and assistance for designing the field tests, and in helping to analyze the results from the tracer test. Two different approaches to modeling the gas flow have been pursued. The first, by assuming homogeneity with respect to permeability, allows solutions to the boundary value problems to be obtained in closed form. The second approach allows permeability as a function of space and, hence, relies upon finite difference methods of solution.

Work this quarter has centered upon completing the development of the two models and applying them to the specific tracer test at NRTS. The most difficult aspect of mathematically predicting the gas flow is found in the proper characterization of the permeability of the field site. For the layered model using finite differences, it was necessary to use larger values of permeability for the most permeable zone of basalt into which the gas was injected than were predicted from analysis of air pumping tests conducted prior to the actual tracer injection. This result is reasonable since the air pumping tests were analyzed to estimate permeability on the basis of a homogeneous model. The most permeable zone of basalt is overlain by a less permeable basalt layer. This less permeable layer would tend to retard the flow in the field during

the air pumping test. Thus, analysis using an assumption of homogeneous media would result in values of permeability smaller than the true value for the most permeable zone.

For the homogeneous model, representation of the minimum time for gas travel back to ground surface was greatly simplified by using approximations which sacrificed not more than about 5% accuracy over the range of injection pressures considered. Methods were also devised for obtaining the flow of gas per unit time per unit area (flux) at the ground surface in both modeling approaches. These results can be compared with flux measurements made in the field during the tracer test.

The writing of the final reports on the modeling approaches was begun during the quarter. These reports, when completed, will include the analysis of the field tracer test.

SIMULATION MODELING OF THERMAL GENERATION IN SELECTED
RIVER SYSTEMS - R. T. Jaske

Operations research with the Col HEAT thermal simulation model currently in use to investigate the effects of future nuclear power plants on temperatures of the Upper Mississippi River Drainage Basin is now well under way. During the report period, the following specific accomplishments were made:

- The results of previous work on the upper Illinois Water Waterway were published in report BNWL-728, "A Test Simulation of the Temperature of the Illinois River and a Prediction of the Effects of Dresden II and Dresden III Reactors," dated April, 1968.
- Additional runs carrying the results of the published document forward from Mile 230 down to the mouth of the Illinois River have been completed for the cases of the previous study. These runs indicate that, even at this extended distance, measurable fractions of the thermal modification at Starved Rock Dam 35 miles below the plant

remain down to the mouth of the River. A follow-up report is in preparation.

- ● Test simulations of the main stem Mississippi River between Minneapolis-St. Paul and Alton, Illinois have been carried out with test data collected earlier in the program. The simulations are quite satisfactory although occasional unexplained deviations are under study. Previous tests of this type have revealed that, in most cases, the computer system is more accurate than the measured data. The latter, in many cases, has not been collected with the objective of use in thermal digital simulation.
- ● Two especially calibrated thermographs were prepared at Battelle-Northwest and shipped to the USGS in Champaign, Illinois, the cooperating office of the Survey. They were installed and checked by Battelle personnel at (1) La Grange Dam at Mile 80 on the Illinois Waterway, and (2) at Dam 28 on the Mississippi River at Alton, Illinois. These instruments will supplement and verify the thermal record at that location and provide a basis for the evaluation of the start-up of the Dresden and Quad-Cities reactors as the units are completed.

The schedule for completion of the Upper Mississippi study now calls for formal completion and publication sometime in September. Steps are now under way to identify other regions of interest to the program of the Division of Reactor Development and Technology. The two proposed areas are:

- ● The Great Lakes region, specifically Lake Michigan.
- ● The Ohio River above the confluence with the Tennessee and the Cumberland, including the entire upper drainage basin.

Model development proceeding concurrently with operations research is directed toward creation of a national thermal modification evaluation system which could serve as the basis

for adjudication, and as a licensing activity by other agencies. Incorporation of the magnetic weather file data of interest to the program is now under study, along with an interface to the Storet FWPCA sponsored water quality data system. A completely revised computer model with improved capability is now operational. This development removed some built-in limitations related to the origin of the system for use in the large flow system of the Columbia River. Improved methods for reducing maps and soundings charts are under study with the objective of developing a volumetric model of the entire national water system in a practicable time period.

5. FIXATION OF RADIOACTIVE RESIDUES

INTRODUCTION

This progress report is the thirty-sixth in a series presenting research and development activities in the field of radioactive wastes.

Experimental work charged to programs other than those of the Division of Reactor Development and Technology is sometimes included for general interest and completeness. Such work is identified in the headings.

PHOSPHATE GLASS SOLIDIFICATION PROCESS TECHNOLOGY

Foaming in Evaporator - J. E. Mendel

Laboratory tests indicated the probable source of the foaming observed during Runs PG-1, 2, and 3 in the WSEP was dibutyl phosphate (DBP), introduced with the Purex Plant waste solution. Commercial silicone antifoam agents successfully counteracted the DBP-induced foaming tendency in a laboratory test apparatus.

The test apparatus was an immersion heater surrounded by a shroud and baffle arrangement designed so that the solution movement in the WSEP evaporator was simulated on a small scale. Initial experiments established that laboratory-prepared PW-2 phosphate glass process feed solution did not foam when the evaporator was operated anywhere in the range of boiling points 125-140 °C. The Purex waste, possibly containing a series of organic contaminants, was then tested with the laboratory evaporator operating at boiling point 130 °C. Also tested was di(2-ethylhexyl) phosphoric acid, the extractant used in fission product recovery.

<u>Contaminant</u>	<u>Result of Foam Test</u>
Degraded NPH	negative
Tributyl phosphate	negative
Dibutyl phosphate	negative
Monobutyl phosphate	negative
di(2-ethylhexyl) phosphoric acid	negative

NPH, the normal paraffin hydrocarbon currently used as diluent in the Purex solvent, was chemically degraded by heating with HNO_3 - HNO_2 .

Dibutyl phosphate, a degradation product of the Purex extractant, tributyl phosphate, was the only contaminant to cause significant foaming. Maximum foaming occurred with about 200 ppm DBP, a concentration at which the foam could not normally be retained in the test apparatus with the 240 W heat input used in all tests. The foaming decreased to negligible amounts at DBP concentrations below 30 and above 1600 ppm.

Two commercial silicone-type of antifoam agents, and one commercial magnesium silicate antifoam agent were tested for their ability to counteract the foaming due to DBP. All tests were made at 130 °C. The silicone antifoam agents suppressed the foam, while the magnesium silicate did not. One silicone antifoam was chosen for longer term testing. Starting with 200 ppm DBP and 100 ppm of antifoam, foaming was suppressed for approximately 2 hr, at which time more antifoam had to be added to prevent foaming out of the apparatus. As the test was continued, a tendency for successive antifoam additions to become less effective was noted. This tendency, however, was not observed in the plant test.

Platinum Melter, Feed Solution, and Condenser Corrosion -

R. F. Maness

Three laboratory-scale phosphate glass runs were made with semicontinuous addition of feed to an alumina crucible equipped

with an overflow weir. A high phosphate feed used in the first run was prepared from a sulfate-free (PW-1) waste solution containing 783 milliliter of 85 wt% H_3PO_4 and 209 g NaNO_3 /liter of PW-1 solution. Feed for the second and third runs was prepared from PW-1 solution containing 406 milliliters of 85 wt% H_3PO_4 and 168 g NaNO_3 /liter of PW solution. In addition, feed for the second run contained 0.01M Pb(II) , while feed for the third run contained 0.5 g of silicone antifoam agent/liter of PW-1 solution. The first and second runs were made at 1000 °C, whereas the third run was made at 1100 °C. Platinum specimens were exposed to the liquid and to the vapor phase. Platinum corrosion in the liquid phase was essentially nil in the run made with high phosphate feed but significant corrosion, 0.3 mils/mo, occurred on the specimen exposed to the vapor phase. In the run made with feeding containing Pb(II) , platinum exposed to the melt corroded at a measurable rate, 0.12 mils/mo, whereas vapor phase corrosion was nil. Both liquid and vapor phase corrosion was nil in the run made with feed containing the antifoam agent. In no case was there evidence of cracking or embrittlement.

Corrosion rates of Ti, 304L, and Hastelloy C* were determined in simulated low salt waste (PW-4) solution boiling at 125 °C. Corrosion rates were <0.1, 34, and 14 mils/mo, respectively, as determined by weight loss after five 24-hr exposures.

Nionel** and Carpenter 20[†] Cb-3 specimens were evaluated for corrosion resistance in HNO_3 - H_2SO_4 - H_3PO_4 systems simulating the condensate from the melter of the phosphate glass process. The condensate composition, when not diluted with steam, is about 10M HNO_3 -5M H_2SO_4 -0.1M H_3PO_4 . This system, at

* Registered trademark of Union Carbide Company.

** Registered trademark of International Nickel Company.

† Registered trademark of the Carpenter Steel Company.

the boiling point, corroded Nionel at a rate of 2.2 mils/mo. A 50% dilution of the system with water reduced the corrosion rate to 0.2 mils/mo. Reducing only the nitric acid concentration, or only the sulfuric acid concentration by a factor of two reduced Nionel corrosion by a factor of two. In all cases, corrosion of Carpenter 20 Cb-3 was higher than that of Nionel by a factor of 1.5 to 2.5.

RADIOACTIVE DEMONSTRATION - J. N. Hartley and J. L. McElroy

Run PG-4

During Run PG-4, wastes equivalent to 1.3 tonnes of 1.6 yr old 20,000 MWd/tonne power reactor fuel were processed to produce 213 kg of glass product. The 73 liters of radioactive glass produced a self-generating heat density of 57 W/liter (4200 W total).

The denitrator was fed 725 liters of adjusted sulfate containing PW-2 type feed (540 liters/tonne) (see Table 5.I for composition) in 77 hr at an average feed rate of 9.4 liters/hr. The feed contained 1,160,000 Ci of radioactivity including 33,000 Ci of radoruthenium. The feed was concentrated in the denitrator-evaporator by a factor of 5.1. The continuous addition of a commercial silicone antifoam agent to the denitrator at a rate of ~100 ppm/hr (3.9 liters solution/hr) and top entry feeding into the vapor space of the denitrator was sufficient to control foaming. Denitrator foaming was allowed to occur prior to starting the antifoam addition. None of the denitrator contents foamed over into the melter condensate. Additional operating parameters and collected data for Runs PG-4 and PG-5 are listed in Table II. Also see Figure 5.1 for Mode A equipment arrangement.

Prior to the run, laboratory investigations on nonradioactive feed (discussed above in "Process Technology") and on actual radioactive feed from the previous Run PG-3 were made to

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FIGURE 5.1. Mode A Arrangement for PG-4

TABLE 5.I. Feed Composition for PG-4 and PG-5

Component	PW-2 for PG-4		PW-1 for PG-5	
	Molarity, M(a)	Lab Analysis, M	Molarity, M(a)	Lab Analysis, M
H ⁺	---	4.3		
Fe ⁺³	0.184		0.93	0.4
Cr ⁺³	0.024		0.012	
Ni ⁺²	0.01		0.005	
Al ⁺³	0.262 ^(b)		0.001	
Na ⁺¹	0.95	0.99	0.138	
UO ₂ ⁺²	0.01		0.010	
NO ₂ -NO ₃ ⁻¹	5.37	4.15	7.5	
SO ₄ ⁻²	0.86	0.97	--	
PO ₄ ⁻³	0.006		0.003	
SiO ₃ ⁻²	0.01		0.01	
Cs ⁺ (added K)	0.42		0.42	
Sr ⁺²	0.0155		0.0155	
Ba ⁺²	0.0195		0.0195	
(Y + Re) ⁺³	0.12		0.12	
ZrO ⁺²	0.065		0.065	
MoO ₄ ⁻² + Tc as MoO ₄ ⁻²	0.079		0.079	
Ru ⁺²	0.0032		0.0032	
Ru ⁺² (added Fe ⁺³)	0.0288		0.0288	
Rh ⁺² (added Co ⁺²)	0.0074		0.0074	
Pd ⁺² (added Ni ⁺²)	0.0172		0.0172	
Ag ⁺² + Cd ⁺² (added Cu ⁺²)	0.0016		0.0016	
Te ⁺⁴ (added SO ₄ ⁻²)	0.0064		0.0064	
<u>Additive to Feed</u>				
Na ⁺¹	---	---	2.46	---
M _T ⁺	3.48		3.77	
PO ₄ ⁻³	3.36	3.2	3.77	---
M _T ⁺ /P		1.09		1.0
Sp gr @ 25 °C		1.43		1.38
% Solids		10% solid 56% slurry		65% slurry
Final Volume, liters/tonne		540		666
<u>Radioactivity</u>				
Total, Ci/liter		1590		3000
Ru, Ci/liter		46		53.4
Heat, W/liter		7		12.1

(a) Molarities are at 378 liters/tonne.

(b) Aluminum is considered as a substitute for part of the iron.

TABLE 5.II. WSEP Phosphate Glass Solidification Operating Parameters and Results

Operational Data		
Run No.	PG-4	PG-5
Date	2/68	4/68
Feed Type	PW-2 (a)	PW-1
Feed, liter/tonne	540	666
Pot Diam., in.	8	12
Pot Material	Mild Steel	Mild Steel
Run Feed Time, hr	77	104
Avg. Feed Rate, liter/hr	9.4	8
Avg. Steady State Feed Rate, liter/hr	7.5	NA (c)
Cooling, hr	2 (b)	NA
Total Feed, liters	725	825
Total Feed, tonne equiv.	1.3	1.2
Feed: Solid Vol. Ratio	9.1	2.1
Operating Mode	A	A,B
Problems	1. Overfed melter causing high level of foam in melter	1. Continuous plugging problems in airlift pot recirculation line, melter feed line, and denitrator dip tubes
Fraction of Activity in Condensate, Total Less Ru and /Ru		
Thru Melter Condenser	$\sim 2 \times 10^{-3} / 1.2 \times 10^{-2}$	$3.2 \times 10^{-3} / 9.9 \times 10^{-2}$
Thru 1st Condenser	$3.5 \times 10^{-3} / 2.2 \times 10^{-2}$	NA/NA
Thru Evap. Condenser	$3.1 \times 10^{-7} / 8.5 \times 10^{-4}$	$4.6 \times 10^{-6} / 2.7 \times 10^{-3}$
Thru Fract. Condenser	$2 \times 10^{-9} / 2 \times 10^{-6}$	$3 \times 10^{-8} / 8.4 \times 10^{-7}$
Off-Gas to Scrubber	$4 \times 10^{-9} / 7 \times 10^{-7}$	$9.4 \times 10^{-10} / 1.6 \times 10^{-6}$
Off-Gas to Stack	$2 \times 10^{-12} / \text{NA}$	$1 \times 10^{-12} / \text{NA}$
Ratio Activity (e)	$4 \times 10^2 / 7.8 \times 10^3$	$1.8 \times 10^3 / 1.8 \times 10^3$
Filled Pot Data		
Total Curies in Feed to Pot	1.2×10^6	2.5×10^6
Curies of Ru in Feed	3.3×10^4	4.4×10^4
Solid in Pot, liters	73.3	115
Solid in Pot, kg	213	400
Solid in Pot, liters/tonne	56.4	95.8
Heat in Pot, W	4200 (d)	8300
Equiv. Aging Time, yr	1.6	0.5
$\bar{C}$ Temp in Cool Furnace, °C	510	740
$\bar{C}$ Temp in Water, °C	NA	NA
Δ Temp $\bar{C}$ to Wall, °C	175	340
Total Corrosion, mm	NA	NA
Welded Lid	Yes	Yes
Pot Self Pressurizer, psi	NA	NA
Pot Leakage, atm cm ³ /sec	NA	NA
Radiation Reading, R/hr @ 6 in.	3.7×10^4	4.7×10^4

(a) Aluminum substituted for ~50% of nominal 0.445 iron

(b) Values are with pot sitting in cool furnace.

(c) Not available

(d) By pot calorimetry data

(e) Ratio of activity concentration in the fractionator condensate to that in 10CFR20

determine the cause of and methods for prevention of foaming. It was established that dibutyl phosphate (DBP) is an effective foaming agent in the evaporator concentrate and was probably the cause of the foaming during previous Runs PG-1, 2, and 3. Silicone antifoam agent was found to counteract the DBP induced foaming in the laboratory test apparatus during short term tests but less successful in tests of several hours duration. The radioactive feed tests were conducted in B-Cell on the feed from PG-3 to investigate foaming in a glass, scaled-down test apparatus similar in geometrical shape to that of the denitrator. Foaming was found to be time-dependent, with some foam-making material accumulating in the denitrator. The continuous addition of the antifoam agent to the system was subsequently found to successfully subdue foam and prevent its recurrence. In the full-scale denitrator, a continuous addition of approximately 100 ppm/hr was required for adequate control.

Startup and shutdown for the denitrator required 12 and 13 hr, respectively, thereby leaving 65 hr of steady-state operation. The average feed rate during steady-state of 7.5 liters/hr was limited by a maximum liquid level of 27 in. in the denitrator. Of the 725 liters of adjusted feed (540 liters/tonne) fed to the denitrator, 669 liters of equivalent feed was airlifted to the melter. After the final dumping of the melter, the pot was cooled to where the steady-state wall temperature in the "cool" furnace would not exceed 425 °C for the required 2 hours. These conditions, yielding an average processing waste rate of 0.35 tonnes of power reactor waste/day, included startup, steady-state operation, shutdown, and cooling time. The steady state processing rate was 0.48 tonnes of power reactor waste/day.

The 57 W/liter of self-generating heat density in the glass was sufficient to produce a pot centerline temperature of 510 °C and a centerline-to-wall ΔT of 175 °C with the

pot in the "cool" furnace. These values indicate effective thermal conductivity of the glass to be 0.44 Btu/(hr)(ft)(°F) [0.76 W/m °C]. The product was a glassy dark greenish-brown color with a measured density of 2.9 kg/liter.

With the melter operating at 1220 °C, the internal melt temperature ranged from 840-1100 °C, depending on feed rate to the melter. 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 receiving pot to completely empty the melter. Receiving pot temperatures were controlled at 550-590 °C to prevent stalagmite formations.

Use of four-year-old radiocerium solution as the main heat contributor for the feed resulted in rare earth content in the feed approximately six times the nominal 0.12M. Additional phosphoric acid and sodium (2.17M and 0.5M, respectively) were added to form a good melt.

Aluminum was substituted for ~50% of the nominal 0.445M iron in the PW-2 feed because of excess aluminum in the sulfate-free waste stock from the Purex Reprocessing Plant. This aluminum produced no noticeable change in the expected processing characteristics.

During PG-4, 2.2% of the total ruthenium and 0.35% of other radioactive species fed to the denitrator were volatilized or entrained. Likewise, 1.3% of the total ruthenium and 0.22% of all other radioactive species fed to the melter were volatilized or entrained. Figure 5.2 shows the fission product accumulation in the melter condensate.

The ruthenium decontamination factor, DF,* (cumulative for the entire run) across the WSEP condensate evaporator,

* *Total curies in evaporator at end of run from any source/total curies gained in fractionator.*

FIGURE 5.2. Fission Product Accumulation
in Melter Condensate (WSEP
Run PG-4)

was 32. The cumulative DF for nonvolatiles across the evaporator was 3.1×10^5 . The fractionator had a DF* for ruthenium and nonvolatiles of 400 and 300, respectively. Figure 5.3 shows the ruthenium volatility from the WSEP evaporator.

The fraction of feed activity in the final condensate stream from the acid fractionator was 2×10^{-6} for ruthenium and 2×10^{-9} for other combined fission products. These fractions represent activity above CFR20 limits** by factors of 7800 and 400.

A test conducted to investigate the effects of air sparging the melter on melter capacity was inconclusive.

Run PG-5

Phosphate Glass Run PG-5 was completed in WSEP using a sulfate-free PW-1 type feed containing 2.46M sodium additive as a melt-making flux. During PG-5, wastes equivalent to 1.2 tonnes of 0.5 yr-old 20,000 MWd/tonne power reactor fuel were processed to produce 315 kg of glass product. The 115 liters of radioactive glass produced a self-generating heat density of 77 W/liter (8800 W total).

The denitrator was fed 825 liters of adjusted sulfate-free PW-1 type feed (666 liters/tonne) in 104 hr at an average feed rate of 8 liters/hour. The composition is shown in Table 5.I. The feed contained 2,500,000 Ci of radioactivity (10,000 W) including 44,000 Ci of radioruthenium. The feed was concentrated in the denitrator-evaporator by a factor of approximately 3. Continuous addition of silicone antifoam agent was used throughout the run at an average rate of 11 g/hr (1-1/2 liters/hr solution). The antifoam addition completely suppressed any foaming in the denitrator.

* Total curies in fractionator at end of run from any source/total curies gained in fractionator condensate.

** AEC's Rules and Regulations, 10-CFR 20, Table II Column II.

FIGURE 5.3. Ruthenium Volatility from WSEP Evaporator
(WSEP Run PG-4)

Approximately 2,000,000 Ci of radioactive waste was concentrated during the run, along with the denitrator condensate in the WSEP evaporator for use during the next run, PG-6. Figure 5.4 shows the Mode B equipment arrangement for performing this operation.

The melter feed line became plugged with solids from the denitrator concentrate after 38 hr of operation. The run was temporarily shut down until the melter feed line was finally unplugged by heating the feed line with 130 °C steam while lightly tapping on the line. The line was then flushed with nitric acid and the run was later restarted. Plugging difficulties with specific gravity and weight factor dip tubes of the denitrator did not occur until approximately 50 hr had elapsed during both parts of the run. Continuous steam purges were used to prevent plugging during this period, and dip tubes were then steam purged once every 2 hr to satisfactorily prevent plugging. Some plugging difficulties in the airlift pot recirculation line to the denitrator were encountered and overcome by the use of the remote features of the newly-installed airlift pot, and by acid sprays of the entire pot. The remote feature used in helping to unplug the recirculation line, the adjustable recirculation plug valve, was extended in and out of the recirculation line.

Excessive foaming, occurring only once in the melter, was generally prevented by maintaining the melt above 950 °C. The rate of radioactive glass discharge from the melter to the 12-in. diameter mild steel pot was approximately 1 liter of glass/hour. With the pot setting in the cool furnace, the 8800 W of internal heat produced a centerline temperature of 700 °C and a centerline-to-wall temperature difference of 375 °C. These values give an effective thermal conductivity of 0.58 Btu/(hr)(ft)(°F). No plugging of the dip tubes

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FIGURE 5.4. Mode B Arrangement for PG-5

tubes occurred in the WSEP auxiliary evaporator in contrast to several previous runs. This lack of plugging was attributed to a thorough flushing of the dip tubes prior to PG-5 by using alternating flushes of caustic and nitric acid.

The only other difficulty occurring during the run was the leak developed by the No. 1 feed pump at 92 hr of feeding. The run was completed using the No. 2 feed pump.

SPRAY SOLIDIFICATION

Spray Calciner Development - W. R. Bond

During the quarter, Runs DSC-15, 16, and 17 were made using a simulated low-salt (PW-4) feed to the spray calciner and platinum melter. Additives used in the first two runs were 1.5M NaNO_3 and 1.1M H_3PO_4 at 100 gal/tonne, while in the third run additives of 1.1M NaPO_3 and 0.4M NaOH at 100 gal/tonne were used. In all three runs the ratio of metal equivalents to phosphorus atoms was 2.3. This formulation had a melting point of 900 °C and a calcine stick point of 750 °C which are compatible with spray calciner and platinum melter operation.

Pertinent run conditions and results of the runs are presented presented in Table 5.III. The calciner barrel after Run DSC-15 was badly caked with a 1/2 to 3/4-in. thick calcine scale the full length of the barrel. The calcine apparently had been wet and, after contacting the hot walls, dried there as a cake. No sintering of the calcine was indicated, and the deposition was apparently the result of exceeding the drying capacity of the calciner. The total of 9 kg of calcine was removed from the calciner barrel. For Runs DSC-16 and 17, lower feed rates (see Table 5.III) were used than in DSC-15. Although inspection of the calciner after Run DSC-16 revealed some scale, less than 2 kg of calcine was removed from the barrel. The calciner barrel was relatively clean after Run DSC-17. No significant deposition occurred in the calciner cone during any of the runs.

TABLE 5.III. Run Conditions and Results

<u>Run No.</u>	<u>DSC-15</u>	<u>DSC-16</u>	<u>DSC-17</u>
Feed Type	PW-4	PW-4	PW-4
Atomizing Gas	Air	Air	Air
Calciner Furnace Temperature, °C	675-700	690-700	700
Melter Furnace Temperature, °C	1000-1025	1025-1060	1025-1110
Pot Furnace Temperature, °C	--	--	800
Feed Consumed, liters	164	174	154
Feed Concentration, liters/tonne	379	454	454
Feed On-Time, hr	9	12	10
Average Feed Rate, liters/hr	17.2 1st half 19.2 2nd half	14.5	15.4
Product Collected, kg	20.9	19.8	18.7
Product Concentration, liters/tonne	27.2	27.4	26.4
Pot Material	304L SS	304L SS	304L SS
Pot Diameter, in.	8	8	8

The filter pressure drop during DSC-15 ranged between 10 and 11 in. of H₂O at a blowback steam pressure of 30 psig. The blowback pressure was increased to 50 psig for one hour midway through the run. The filter pressure drop was reduced to 7 in. of H₂O during this period, and chunks of filter coat were noted falling into the melter. Pressure drop across the filter rose again to the previous range after the blowback pressure was reduced to 30 psig. Inspection of the filters after the run revealed the bottom half of the filters to be relatively clean and the filter coat to be approximately 1/16 in. thick. The top section of filters displayed a very dense filter coat, 1/8 to 1/4 in. thick, and large chunks of

calcine lodged between the filters. Apparently chunks of the filter coat broken loose during the use of 50 psig blowback steam were unable to fall clear of the filter because of their size. The application of 90 psig blowback air failed to remove any calcine from the filters. The filters were cleaned in-place by injecting nitric acid directly into the blowback steam.

During the first five hours of Run DSC-16, pressure drop across the filter rose from 0.5 to 8 in. of H_2O with the use of 30 psig blowback steam. An increase in blowback pressure to 40 psig was required to level the filter pressure drop out at 8.5 in. of H_2O . Inspection of the filters after the run revealed them to be covered with a dense filter coat from 1/8 to 3/16 in. thick. There was no holdup of calcine between the filters. The application of 90 psig blowback air reduced the filter coat to less than 1/16 in. thick. The filters were then cleaned by back flushing with water.

Considerable difficulty was experienced with excessive filter pressure drop during Run DSC-17. The pressure drop across the filter rose from 0.5 to 2.5 of H_2O during the 15 min that water was fed, and rose again to 7.0 in. of H_2O within 5 min after the feed was turned on. Despite raising the blowback steam pressure progressively from 35 to 65 psig and reducing the blowback cycle from 2 to 1 min, the filter pressure drop increased to 15 in. of H_2O after 7 hr of feeding. The feed and atomizing air were turned off at this point and the filters were blown back with 70 psig steam for 20 min. Restart of the run revealed a reduction in the filter pressure drop to only 12 in. of H_2O , and an increase to 15.3 in. of H_2O at the end of the run despite the use of 70 psig blowback steam. Post-run inspection revealed the filters to be coated, as in the previous runs, with a very dense coat of fine calcine approximately 3/16 in. thick.

The PW-4 melt was discharged batchwise from the melter via the freeze valve during both DSC-15 and 16. Melt batches ranged from 2 to 4 liters and were discharged in one to two minutes. The dump procedure required reducing the calciner vacuum to 1 to 2 in. of H_2O just prior to the dump so as to essentially empty the melter. The calciner vacuum was increased back to normal immediately after the dump and, in some cases, reduced again briefly to reseal the freeze valve. The calciner vacuum during one dump was not reduced. The melter continued to drip after the dump, and complete emptying of the melter by reducing the calciner vacuum was required in order to reseal the freeze valve. The drip discharge of the melter at the end of this dump was erratic and caused partial blockage of the freeze valve port. In general, the melter drained during a dump in a straight, steady stream. One exception occurred when the freeze valve thermocouple, protruding into the melt stream, created a stalagmite completely filling the freeze valve port. Although the receiver pot was unheated during both these runs, the melt dumps filled the pot evenly. Sectioning of the pot revealed some void space between the batches, and gas pockets within the batches. The bulk density of the product as measured in the receiver was 2.6 kg/liter, or approximately 83% of theoretical.

The melt was discharged continuously during DSC-17 from the melter via the weir which drained both dropwise and in a steady stream as a result of pressure fluctuations in the melter caused by the filter blowback. Immediately following a pressure increase in the melter, the steady stream melt discharge would become quite erratic just prior to a dropwise discharge transition and, as a result, considerable melt would splatter against the sides of the weir port to form stalactites. Several of these had to be broken off during the run and one almost completely filled the weir port.

The weir discharge ceased 1-1/2 hr before the end of the run as a drop of melt solidified just off the end of the weir tip. It was necessary at the end of the run to raise the freeze valve temperature to 1020 °C before the melt could be drained from the melter. Discharge from the freeze valve ceased with about a 1/2 liter of melt remaining in the melter, and a drop of melt was also found to be solidified off its tip. The drip point of the melt, determined to be 860 °C, was comparable to that of previous runs. There was some splatter on the walls and thermowell of the receiver pot as a result of the erratic weir discharge but no gross deposition. The bulk of the melt had spread out evenly in the bottom of the heated receiver pot.

Considerable denitration of the calcine was noted in the melter during the first two runs. Analyses of calcine samples indicated that calcine from DSC-15 contained as much as 18 wt% nitrate, and calcine from DSC-16 contained as much as 10 wt%. The nitrate contents of PW-1 and PW-2 calcines were routinely less than 2 wt% under similar operating conditions. Analytical data on ruthenium volatility during Run DSC-16 indicated instantaneous losses of ruthenium to the condensate in the range of 4 to 20% during the run, with an overall loss of 12%. However, analyses of calcine and melt samples indicated losses of ruthenium to range from 80 to 90%. This discrepancy has not been resolved. The effect on the calcine denitration and ruthenium volatility, as a result of using NaPO_3 and NaOH as additives during Run DSC-17, instead of H_3PO_4 and NaNO_3 , is not known as yet due to the unavailability of analytical results.

Melt Container Corrosion - R. F. Maness

A simulated spray-solidified phosphate melt prepared from PW-4 solution ($\text{Meq/P} = 2.3$) corroded both 304-L and 310 stainless steel at a rate of about 15 mils/mo in 100 hr tests at 800 °C.

PRODUCT CHARACTERIZATION AND STORAGE - V. P. Kelly

A-Cell Modifications for SSETF

Beneficial occupancy of A-Cell for the Solids Storage Engineering Test Facility (SSETF) was achieved as scheduled with the completion of in-cell modifications by mid-March. Completed items included installation of the second floor window and the stainless-steel wall liner. Items remaining to complete the project are design and fabrication of a wall transfer mechanism, fabrication of a shielded ion chamber, and completion of miscellaneous gallery items.

SSETF Equipment and Systems Fabrication and Installation

Fabrication and installation of SSETF equipment proceeded at a vigorous pace. Items of equipment include:

- The pot storage rack, a stainless-steel pipe structure to be installed in the cell to support test pods and pots during assembly.
- Sixteen test pods (10 completely assembled).
- The third-floor TV plug required for viewing upper level remote operations.
- The remotely-operated mirror unit for additional viewing aid.
- Electrical, instrument, mechanical drive, welding, leak test, and air supply wall plugs.
- Three test pod heater control units, providing a total of seven of the required 10 units.
- The 300-point digital data acquisition system for temperature monitoring (about 50% complete).

Pot transfer hardware and incell tool systems are required to activate the cell.

Remote Handling Tests

Remote handling design verification testing is required to verify operability of in-cell components. The first phase of these tests was completed with the transfer of four storage pods from the fabrication shop to the cell, and remote installation of the first pod in a second-floor cubicle opening. Installation of handling pods into third-floor cubicle openings, removal of lights and filters, and assembly of a pot and pod will complete remote handling tests.

LABORATORY STUDIES

PG-1 and PG-2 Leach Rate Measurements - J. E. Mendel

Leach rates on a grab sample of phosphate glass product from WSEP Run PG-1 have been measured in room-temperature water on a weekly basis for 4-1/2 months. The leach rate, based on the amount of ^{137}Cs found in the leach water, has leveled off at about $9.0 \times 10^{-7} \text{ gm/cm}^2/\text{day}$. The leach rate, based on ^{144}Ce , is about $1.8 \times 10^{-7} \text{ gm/cm}^2/\text{day}$.

A grab sample of phosphate glass product from WSEP Run PG-2 has been leached 2-3/4 months. The leach rates are 1.4×10^{-6} and $2.4 \times 10^{-7} \text{ gm/cm}^2/\text{day}$, based on ^{137}Cs and ^{144}Ce , respectively, and may still be decreasing slowly with time. The higher activity in Run PG-2 enabled $^{95}\text{ZrNB}$ and ^{106}Ru also to be detected in the leach water. The leaching of $^{95}\text{ZrNB}$ at about the same rate as ^{137}Cs was determined, while ^{106}Ru leached at a lower rate, approximating that of ^{144}Ce .

Study of Molten Core Storage of Solidified Waste Containing Sulfate - D. W. Brown

Decomposition of sulfate gas resulting in pressure buildup is most probable under high-temperature conditions established for the molten core storage of melt-solidified waste products high in sulfate content. A vessel has been designed and built whereby molten core storage can be simulated, and pressure buildup due to sulfate decomposition can be monitored.

Installation will be accomplished by centering a heater in a 12-in. diameter storage pot about 30-in. high. The storage pot will be filled with simulated spray solidifier sulfate-containing PW-2 waste. The experiment will begin soon.

INTERMEDIATE AND LOW LEVEL WASTE

W. T. McKean

All major components for the Distillation Demonstration Unit to be used for pilot scale distillation of melter condensate from the phosphate glass process have been received. Exceptions are the tantalum tank and piping and the Sihi* vacuum pump. Installation of vessels, piping, and instruments is in progress.

Preliminary work using laboratory-scale apparatus similar to that at BNL is proceeding in C-Cell. Preliminary results indicate that ruthenium evolution can be minimized by using semi-continuous operation and nitrogen sparge. BNL efforts to determine the effect of acid ratios and concentrations on the decontamination factors are currently under way.

The results of the laboratory scale runs will be used to direct tests with the Distillation Demonstration Unit. Tentatively nitrogen sparge will be employed and semicontinuous feed operations will be tested.

SUPPORTING ACTIVITIES - D. W. Brown

Preliminary investigation of methods for reducing ruthenium volatility, or of removing ruthenium from the off-gas to acceptable waste solidification levels has been started. One trap, an electrical capacitor type arrangement 2 in. long by 1/2 in. diam with one lead connecting to a wool plug and the other lead wrapped around the glass, was tested. This run was conducted at 115 °C, 1500 V, and zero current across the trap. About 5% of the ruthenium input was obtained past the trap

* A product of Heraeus-Engelhard Vacuum, Inc.

compared to about 20% of the ruthenium input past the trap with 30 V and a bed temperature of 170 °C. This approach should be investigated further at higher potentials and at varying bed temperatures, with tracer ruthenium employed for analytical purposes.

Formaldehyde additions to the feed lowered the ruthenium volatilized to 4.3%, as compared to 37% ruthenium volatilized without the use of formaldehyde. However, formaldehyde is too reactive for easy handling and a large quantity is apparently needed. Formaldehyde in the amount of 18.5% by volume was added to the feed solution to effect this decrease.

A more satisfactory additive was hypophosphorus acid, H_3PO_2 . When 1M H_3PO_2 and 0.097M cold ruthenium were used in PW-1 spray solidifier feed, the amount of ruthenium volatilized was less than the minimum detection limit of 2.75% compared to 37% volatilized when no H_3PO_2 was used. On heating PW-1 spray solidifier feed solution containing 1M H_3PO_2 , visible NO_3^- decomposition started at about 80 °C. Analyses indicate 0.25M H_3PO_2 to have little effect on Ru volatility with PW-2 (without sulfate), spray solidifier, or pot calcination feeds.

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