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PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
FOR JUNE 1967

AEC DIVISION OF
REACTOR DEVELOPMENT AND TECHNOLOGY PROGRAMS

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By
The Staff of Battelle-Northwest
S. L. Fawcett, Director

July 1967

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

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SUMMARY

CIVILIAN POWER REACTORSNuclear Systems and Concepts Analysis

Revised fuel-cost-area electrical demands, fossil fuel costs, and capital costs have been completed for the final Phase II demonstration. Computer production runs are being made for concluding Phase II and III of the Task Force requirements.

Seventeen RBU Monte Carlo problems have been submitted, and the calculations on 12 have been completed. This study is using BNWL's latest cross section data sets and the most recent version of Monte Carlo which produces an auxiliary output tape for statistical analysis of the results. Two minor changes were made for this study to improve the speed in these small cells.

Credits of \$10/g for U-236, \$50/g for Np-237, and \$150/g for Pu-238 in a D₂O lattice were applied to the fuel costs for thorium systems enriched with U-235, decreasing in the minimized fuel cycle costs from 1.14 to 0.930 mills/kWh_e for the thorium metal driven at 60 W/g at a fabrication rate of \$40/kg.

Thorium loadings with various enrichments of U-233 and U-235 are being analyzed (for full time power operation) with the computer code chain JASON-ALTHAEA-QUICK. Previous burnup results using ALTHAEA for NPR natural uranium loadings will be compared with actual burnup data to assure ALTHAEA's accuracy for various other loadings.

A search of Nuclear Science Abstracts seeking thermal scattering cross section data for H₂O and D₂O did not indicate the existence of any more recent formulae than we are presently using.

All versions of the Fuel Cycle Analysis chain have been converted to the 1108. The execution time on the 1108 is about four times faster than observed on the 1107.

The JASON code now prints out more of the input data than previously and makes more output available to ALTHAEA.

Conceptual Reactor Design Studies

Gross heat capacity calculations have been completed for the major streams in the Lower North Atlantic Slope Basins, New York to York River.

An improvement in the burnup calculations has led to better performance for the steam cooled breeders.

From this past month's results, it appears that pin diameter may vary from 0.15 to 0.40 inch without any appreciable effect on the reactor physics. Heavy steam gives sufficiently increased breeding ratios in the wetter cases to be of further interest. The burnup runs and shape factor curves will have to be rerun with ORNL cross sections in order to be consistent.

The results of the parametric physics study have been incorporated into the design and economic simulations of the SCFBR. A preliminary parametric study based on the Karlsruhe 2500 psi design was presented to the Alternate Coolant Task Force meeting at ORNL on June 21 and 22. Some changes have been made to the design simulation to match the Karlsruhe design and more will need to be made to permit simulating designs at steam pressures below 1800 psi. The fuel cycle unit costs are being examined to provide results more consistent with those of ORNL. The clad design procedure will require modifications in order to increase its range of applicability.

USAEC-AECL Cooperative Program

Analytical studies and laboratory experiments to determine the amount of cross-channel mixing during boiling in rod bundle fuel elements were completed. A report describing the analytical studies was issued earlier (BNWL-371 Pt 1). Two additional reports (BNWL-371 Pt 2 and Pt 3) describing the laboratory experiments are currently under preparation.

The construction of an electrically-heated test section to study the effects of rod spacing on boiling burnout in rod bundle fuel elements is being completed. This test section will allow the study of the effects of both adjacent heated rods and unheated surfaces at spacings between 0.015 to 0.080 inch on boiling burnout. Experiments on this test section are scheduled for FY-68.

Fatigue tests on heat treated and aged Zr-Nb alloy tubing show that the crack length at which leakage occurs is a function of tube geometry and internal pressure. These tests also suggest that the critical crack length for catastrophic failure decreases with increased amounts of fatigue damage.

The analog computer study of hydraulic and nuclear stability of the BLW-250 has been completed. A rough draft of the final report has been written and is nearly ready for publication as a formal report.

An in-reactor stress rupture test conducted at 400 C and 38.7 kg-mm⁻² (55,000 psi) stress ruptured in 156 hrs with 23% total elongation. Control creep tests at 400°C resulted in a similar amount of elongation.

Crud deposits on coupons of six zirconium alloys were measured after 174 days in-reactor in pH-10 NH₄OH at 270-280°C. Deposition did not show marked dependence on surface condition and alloy; deposition did vary widely with loop position.

HTGR Graphite

The experimental apparatus for studying the water vapor - graphite reaction in a gamma field was modified to eliminate possible contamination of the gas stream. These modifications permit a more accurate determination of the effect of radiation on the water vapor - graphite reaction.

An electron microscope investigation of the oxidation of TSX graphite by water vapor indicated that the oxidation was qualitatively similar to the thermal reaction between TSX graphite and oxygen.

The water injection test in the DRAGON Reactor was simulated by using a graphite oxidation computer program. The calculated results show that radiation-induced reactions are required to obtain as much CO₂ as was observed in the test.

APPLIED AND REACTOR PHYSICS

Plutonium Criticality Studies

A series of pulsed neutron source experiments have been performed on heterogeneous plutonium systems made up of layers of PuO_2 -polystyrene and Lucite. The neutron decay constant was measured and reactivity determined as a function of assembly height during the approach-to-criticality. The measurements covered a range of subcritical reactivities extending down to -4.5 dollars.

The value of the limiting critical enrichment (smallest uranium enrichment that can be made critical) for homogeneous hydrogen moderated mixtures of uranyl nitrate, $\text{UO}_2(\text{NO}_3)_2$ has been determined as 2.1 wt% with 99% confidence limits being ± 0.03 wt%. Based on these results the single parameter limit for nuclear criticality safety of uranyl nitrate solutions will be 2.07 wt%.

Phoenix Fuel Reactor Program

Analysis of the kinetics measurements made in the CAF experiments is continuing. A paper entitled, "Approach-to-Critical Experiments with Phoenix Fuel," was presented at the 1967 Annual Meeting of the American Nuclear Society held in San Diego, California, on June 12-15, 1967.

The CAF-Phoenix experiments are to be used as calculational "bench marks" to evaluate both computational methods and cross sections for future calculations.

Analysis of the early stages of the PRCF-Phoenix experiment is under way. Current emphasis is on determination of the "as built" configuration and compositions. Measured values of the power peaking at the core reflector interface are lower than the calculated values.

The measured power distributions have been obtained from the MTR-Phoenix fuel mockup experiment in the Plutonium Recycle Critical Facility (PRCF). Measurements were also made of the effect of tapered fuel plates on the core edge power peaking.

The effectiveness of the regulating rod in the MTR mockup is being studied. Preliminary measurements indicate a low reactivity worth.

The temperature coefficient for the PRCF Phoenix fuel experiment was measured to be -1.9% /°C in the temperature range of 30 to 40 °C.

A preliminary thermal hydraulic analysis of a Phoenix core in the MTR has been completed. The results and a description of methods used was presented to MTR personnel.

The proposed flux monitor system was reviewed with MTR personnel, and an order was placed for two mockup fuel boxes containing simulated wands for hydraulic testing in the hydraulic test facility at Idaho Falls.

New Al-Pu fuel plates and new hardware are being prepared for an MTR irradiated experiment.

A coordination meeting on the MTR-Phoenix fuel burnup experiment was held at Idaho Falls on June 27, 1967.

A meeting was held in San Diego on June 14, 1967, to discuss the possible application of Phoenix fuel in HFIR-type cores.

Analytical studies of the physics characteristics of Phoenix fuel loadings in the CNSG have continued.

High Temperature Reactor Physics Studies

The vertical safety rods, horizontal control rods, and the oscillators were accepted by the HTLTR Operations and Systems Unit.

The GAS CYCLE-HEAT design tests were begun and largely completed by the end of the month. The reactor temperature was 550 °C at the end of the month.

Project CAH-100, High Temperature Lattice Test Reactor (HTLTR), was closed out at the end of the month, with a few exceptions whose cost will be accrued against the project authorization.

The reactor physics program is under review in an attempt to obtain data relevant to large HTGR reactors about one year earlier than had previously been planned.

REACTOR FUELS AND MATERIALS

Fast Fuels Oxides and Nitrides

Two experimental mixed nitride fuel capsules (GEH-14-745 and 746) continue to operate satisfactorily in the MTR after accumulating 16,500 MWd/tonne exposure.

A computer study indicates that large voids in the pellet-to-clad sodium bond in these capsules can be tolerated without adversely affecting the safety or value of the experiment.

Preliminary experiments indicate that a single-phase nitride (or carbonitride) can be obtained by carbothermic reduction of uranium-plutonium oxides under flowing nitrogen at 1550-1600 °C.

Metallographic examinations of sodium- and helium-bonded, stainless steel-clad, UN - 20 wt% PuN test capsules, heated for 100 hours at 650 °C, showed no apparent reactions.

Metallographic examination of sodium-bonded, stainless steel-clad, UO₂ - 25 wt% PuO₂ test capsules heat cycled at 95-105 °C for 68 hours (285 cycles) showed no apparent material reactions.

Leaks in the vibratory ball mill jars were repaired with epoxy resin. Stainless steel ball mill jars and balls were unsatisfactory for milling mixed nitrides because of excessive packing of the powders. Surface area equipment was made available for use in evaluating ball milling effectiveness. Operating difficulties with the sintering furnace were believed to be caused by air leakage and by vacuum-pump oil in the sintering chamber.

Basic Swelling Studies

Three capsules continue to operate successfully, and construction is complete on three others.

Density measurements on specimens recovered from capsule P-15 (700 °C, 1000 psi, 0.2-0.8 at.% BU) yielded swelling of 1 to 17% with corresponding "R" (% swelling/at.% BU) values of 5 to 21.

Uranium specimens are being postirradiation annealed at 15,000 psi and 900 °C.

Nondestructive Testing

Development of circuits for use in demonstrating the application of multiparameter test principles to a pulsed eddy current test is continuing. Construction of the multichannel eddy current tester is nearing completion, and a laboratory evaluation of performance has begun.

A study to investigate the theoretical and practical aspects of eddy current sensing coil design is continuing. Experiments to investigate the effect of tuning on probe sensitivity indicated that the fractional impedance change from a drilled hole standard was increased by a factor of 2 to 5 when the coil was tuned to resonance. In other words, significant sensitivity increases can be obtained by operating the probe coil at resonance.

The Kautz function analysis program can now correctly calculate the first pair of Kautz functions to six significant figures. This accurate reproduction stimulates confidence in applying this method of analysis to actual ultrasonic pulse data. Initial errors in the EDPM program have been corrected, and the program now appears to be functioning properly, although debugging of program subroutines is still in progress.

Our Barnes research radiometer, for use in the infrared and thermal research program, has been returned from the manufacturer after correction of faulty internal controller circuitry. Additional testing was conducted using this radiometer in the remote transient infrared testing system. These tests indicated that the performance of the radiometer was substantially improved. Considerable progress was achieved in the development of a wide angle infrared imaging device which has a $20^\circ \times 20^\circ$ field of view, and this system is now about 80% completed.

Nuclear Ceramics

Single-crystal UO_2 specimens were prepared and exchanged for use in basic studies and information exchanges.

Promising results were obtained for nitrogen analysis in uranium nitride by the Kjeldahl fusion method.

A first attempt to prepare UN by reaction of ammonia with UF_4 (at 500-800 $^\circ\text{C}$) did not succeed.

Installation of an electron beam zone-melting unit was completed, and preliminary attempts were made to float-zone melt UO_2 .

Nuclear Graphite

Property measurements were completed on samples from BG-2 and BG-3 EBR-II irradiation pins. Approval is being sought to irradiate an entire 19-rod subassembly in the core of EBR-II.

The last graphite irradiation capsule in the nuclear graphite irradiation series, scheduled for discharge in July, continues to operate satisfactorily at design temperature.

All graphite samples irradiated in the "Proof Test" program showed the sequence contraction-turnaround-growth. This turnaround behavior occurred at a lower exposure for the transverse samples. Data points from all samples of a given orientation clustered together into two separable bands, one for irradiations at 400-800 $^\circ\text{C}$, and the other for irradiations at higher temperatures. Apparently there is a rapid increase in damage rate over the interval 800-900 $^\circ\text{C}$.

Charging of the first capsule in the DFR has been delayed to July 5, due to operating problems at the reactor.

The thermal conductivity of irradiated graphite decreases when the sample is oxidized in air. A 20% loss of weight due to oxidation produces a 50% decrease in the thermal conductivity.

An electron microscope examination of attack on graphite by microwave-excited oxygen reveals a strong dependency on the distance from the discharge region. The rate of attack on graphite downstream from the discharge region is much lower than that on graphite inside the discharge region. The structural features produced from the two attacks are also significantly different.

Fast Reactor Dosimetry and Damage Analyses

Monitor data were analyzed from the second low-power dosimetry experiment conducted in the EBR-II. Total flux was calculated from several different monitors at axial and radial core positions.

Irradiation Damage to Reactor Metals

A high strength iron base alloy Haynes No. 561 and a family of binary vanadium-chromium alloys have been added to the alloy selection screening studies.

Liquid metal capsule GEH-22-4 has been discharged from the ETR for malfunction of the lead tube. GEH-22-5 containing nickel-base alloys is in final assembly and was shipped to NRTS during the week of June 26.

An in-reactor creep test was conducted on hot swaged, sintered molybdenum at 580 °C and 21.1 Kg-mm^{-2} (30,000 psi) stress. There was no difference in creep rate with respect to the control specimen; however, the rupture life was appreciably shorter.

Transmission electron microscopy of an as-irradiated AISI 304 stainless steel tensile specimen revealed defect clusters 50-100 Å in diameter and having a density of about $1.5 \times 10^6/\text{cm}^3$. After testing at 400 °C, the clusters had increased in size, dislocation loops were resolved, and decreased in density. Radiation hardening of the yield strength was observed at 600 °C, but similar hardening of the ultimate tensile strength was significant only between 200 and 400 °C test temperatures.

Stainless steel materials have been given various thermal/mechanical treatments in preparation for an irradiation program. Mockup of the sheath capsules was evaluated. The indicated temperatures in the sheath capsules based on thermal monitors were

found to agree with the predicted temperatures from the STAC computer calculations.

A paper entitled, "The Influence of Thermomechanical Treatments on the Strength, High Temperature Stability, and Microstructure of Hastelloy X-280," by I. S. Levy, was accepted for presentation at, and publication by, the International Conference on the Strength of Metals and Alloys to be held in Tokyo in September 1967.

Analysis of data from pretreated Inconel 600 specimens irradiated at 1250 °F (677 °C) and their controls, after tensile testing at 1350 °F (732 °C) showed that thermal exposure alone resulted in significant hardening and ductility loss. Metallography showed this to be due to precipitation during exposure. Some experimental treatments showed better thermal stability and a 5-fold increase in postirradiation ductility compared to the standard treatment. Metallography showed this to be due to the fact that the pretreatments had already caused significant precipitation prior to exposure so that less occurred during exposure. Irradiation appeared to reduce ductility by grain boundary embrittlement, since yield strengths were lowered.

Since the last reporting period, no other laboratories participating in the intercalibration test program have reported any counting results. Flux for the MTR VH-2 Facility has been calculated using spectral-averaged cross sections for all monitors sent to participants.

Specimens of AISI 304 and 348 stainless steel in both the annealed and cold-worked conditions have been irradiated in the EBR-II to 1.7×10^{22} n/cm². Tensile testing of these specimens is 75% complete.

The isochronal hydrogen evolution characteristics of AISI 304 and 316 stainless steel have been determined for selected test and hydrogen charging conditions. The observed maximum hydrogen evolution rates were greater and occurred at a lower temperature for the AISI 304 stainless steel.

The investigation of the corrosion film on the 304 and 316 stainless steel specimens subjected to 1060 °F (571 °C) sodium for 609 hours in the isothermal loop is complete. The results obtained do not make it clear how the corrosion film was formed.

A sodium metering station has been assembled and tested which will measure volumes of high purity sodium to closed receiving vessels. Analytical samples may be taken for a check on the purity of the supplied sodium. Design for renovation of Building 321-A has advanced to the point of the first round of comment prints. The support system for the sodium loop was loaded for shipment by rail on June 21. A temporary facility for the disposal of sodium by

reaction with water has been set up. Safety approval was obtained, and Fire Department supervision was arranged for the procedure as set up.

Repairs to the ATR model gas loop are being made during this period. An addition is being fabricated to extend the capability of the horizontal test section to allow insertion and withdrawal of the test specimens during high temperature operation.

In-flux weight gains and hydrogen pickups are reported for titanium, niobium, and Zircaloy-2 after a 4-cycle (84-day) G-7 loop exposure.

A paper entitled "The Analysis of Thermally Activated Flow in Alpha Iron Using Temperature Change Techniques," has been prepared for submission to Acta Metallurgica.

Preirradiation testing of the high pressure X-ray cell is complete, and results confirm a previous theoretical analysis. A cubic to monoclinic transformation in Gd_2O_3 has been found at 22 °C and 40 kbars. High pressure polymorphs of Zr, InTe, PbF_2 , SiO_2 , and P, which remain in a metastable state at ambient conditions, have been prepared and are ready for irradiation.

ATR Gas Loop Operation and Maintenance

Review of vendor data for the components of the ATR gas loop continued. Additional research work on the analytical instrumentation is being completed. Surveillance programs are being formulated to determine life expectancy of loop components.

Metallic Fuels Development

Three Th-2.5 wt% U-1.0 wt% Zr fuel elements continue their irradiation in the P-7 hot water loop at the ETR. Exposures range from 9400 to 18,600 MWd/tonne with corresponding volume increases from 1.3 to 3.4%.

Hollow core uranium fuel elements have been successfully irradiated in the ETR M-3 hot water loop to a maximum exposure of 5143 MWd/tonne, and all elements thus far irradiated continue to show slight volume decreases. Neutron radiographic examination of eight elements shows little decrease in the diameter of the central voids up to exposures of 2674 MWd/tonne. Transverse cracks in the uranium fuel detected by neutron radiography at two different irradiation exposures show no evidence of healing during irradiation.

A highly successful extrusion experiment demonstrated the feasibility of directly coextruding 0.600 inch diameter Zircaloy-2 clad rods with central voids as small as 1/8 inch.

ENGINEERING DEVELOPMENT

Neutron Flux Monitors

Test assembly fabrication for the improved U-234 - U-235 regenerative thermal neutron flux detectors and other detector types progressed following acquisition of all ten sensors and specific capsule assembly components.

Microwave and Infrared Detection of Coolant Impurities and Measurement of In-Reactor Temperatures

Successful laboratory operation to 1000 °C was achieved with one experimental microwave assembly being developed for use in measuring reactor in-core high temperatures.

Upstream Boiling Burnout

Analysis of data obtained earlier in this program was continued, and a tentative mechanism was developed that explains phenomena observed during the occurrence of upstream boiling burnout.

UO₂-PuO₂ Fuel Cycles For Fast Reactors

Length of fuel exposure in a fast reactor will have a profound effect on the doubling time for a fast reactor when the out-of-reactor inventory is considered. Curves are given of simplified cases (holding breeding ratios and plant factors constant) to illustrate the effect of exposure on breeding ratios.

PLUTONIUM UTILIZATION PROGRAM

Fuels Development

Work has been initiated to adapt a Lawrence Radiation Laboratory computer program for use in the high exposure plutonium study.

The underwater profilometer was delivered and assembled in the 308 Building for checkout and inspection of nonirradiated fuel rods.

The vacuum fuel rod welding chamber and associated vacuum system have been accepted from the seller.

Structural analysis results of a PRTR fuel rod irradiated at a maximum power of 18.6 kW/ft agree well with the calculated conditions

for this power generation. Irradiation of the hot-pressed UO_2 - PuO_2 19-rod cluster element is progressing satisfactorily at a maximum power generation of 21.5 kW/ft.

The PRTR prototype UO_2 defect test is operating satisfactorily in the ETR P-7 loop. Maximum calculated rod power is 29 kW/ft. The molten radius is calculated to be about 63% of fuel radius.

In an effort to avoid a delay in the proposed ANL-BNW joint program for transient testing thermal reactor oxide fuel rods, a simpler, less expensive, autoclave design is being proposed by BNW.

A study to determine minimum fuel enrichment requirements in subsequent test rods was completed. The results of the study indicate that fuel rods enriched with 5 at.% U-235 (or equivalent Pu content) will provide the maximum energy levels desired and also provide a self-shielding that is not too different from that encountered in commercial power reactor fuels. Recent transient experiments with vibrationally compacted UO_2 (5.01 at.% U-235) fuel rods in the transparent autoclave have confirmed the results of the study.

Four fuel rods instrumented to measure fuel rod gas temperature and pressure are operating satisfactorily in PRTR. The fuel rod gas pressure buildup appears to follow the expected trend.

Reactor Physics

Americium and curium isotopic concentration data from a highly exposed plutonium-aluminum sample irradiated in the PRTR has been received from the Analytical Chemistry Section.

Concentration measurements to determine the amount of B-10 in samples taken from the moderator of PRTR batch core experiment were made in the Thermal Test Reactor.

Analysis of the batch core power tests in the PRTR is in progress.

Calculations have been performed to estimate the tube power for a 19-rod UO_2 - 2 wt% PuO_2 cluster in H_2O located in channel 1550, as a function of coolant temperature.

Gamma scanning of rods received from the EBWR has begun. The power distribution of the first rod scanned is skewed towards the lower half of the rod as expected.

Fabrication of 280 PRCF fuel rods was completed to meet FY-1967 programmatic needs of the program. Commercial procurement to obtain the balance of the (4 wt%) PuO_2 - UO_2 fuel rods during FY-1968 is now being completed under the two-step procurement procedure.

A document entitled "Critical Experiments with Batch Core Fuel in the Plutonium Recycle Critical Facility," has been written.

Three papers were presented at the 1967 Annual Meeting of the American Nuclear Society held in San Diego, California, June 12-15. The titles of the papers were: 1) "Critical Experiments with the UO_2 -2 wt% PuO_2 Batch Core in PRTR," 2) "Experiments and Calculations for H_2O Moderated Assemblies Containing UO_2 -2 wt% Fuel Rods," and 3) "Analysis of a UO_2 19-Rod Cluster Experiment with the RBU Monte Carlo code.

Modifications to the induction plasma arc equipment for spheroidizing PuO_2 were completed.

A set of fuel rods containing solid solution UO_2 -2 wt% PuO_2 (24% Pu-240) was completed.

Analysis of foil activation data is under way to calculate the magnitude of the decrease in k_∞ with increasing PuO_2 particle sizes through the range 0, 100, 200, 350 microns.

Resonance region data for 10 isotopes were updated on the BNW Master Library and Legendre scattering coefficients for seven isotopes were prepared for the ENDF/B library. A new HRG Library tape was prepared. Revisions were made in BARNs-II and in BCDRD to improve efficiency. Conversion of codes from the Univac 1107 to the 1108 was completed.

Program GASKET has been compiled and is running on the Univac 1108. Various other programs were converted from the 1107 to the 1108 computing system.

Reactor Engineering Development

Documentation of the thermal hydraulics design code, REPP, is nearly complete. Development of a detailed over-all design and analysis code has begun.

Materials Development

No evidence of unusual corrosion of aluminum by D_2O with 20 ppm boron was found on examination of corrosion coupons suspended in the moderator for up to 701 days.

The hoop strength versus temperature curve for a $1\frac{1}{2}$ -inch flaw length has been completed from room temperature out of 300 °C. The slope of the curve is approximately -10 psi/°C.

A tentative specification for detection limits for inspections of PRTR process tubes was prepared.

Cycle Analysis

Seventeen RBU Monte Carlo problems have been submitted and the calculations on 12 have been completed, but analysis of the results has not been started. This study is using BNWL's latest cross section data and a most recent version of Monte Carlo which produces an auxiliary output tape for statistical analysis of the results. Two minor changes were made for this study to improve the speed for these small cells.

Test Reactor Operation

The PRTR operated to 64 MW to produce 608 MWd during June. Reactor power was limited to a specific rbd power of 19.7 kw/ft for the average of Ring One fuel elements.

The total output from the PRTR is 1362 MWd of which 1162 MWd have been accumulated by the 55 fuel element batch core.

Three carbon steel elbows, on the steam generator secondary side vent lines, failed by severe localized corrosion attack. The heat exchanger and associated piping were inspected by visual and and radiographic techniques, and no other problem areas were found. New carbon steel elbows were installed, and the system returned to service.

The Fuel Element Rupture Test Facility was removed from irradiation service after ion exchange resin was backwashed into the FERTF coolant.

"Crud" levels continued to be low in the primary coolant system under pH-7 operation.

Seventeen process tubes were examined. There were no significant changes observed since the previous examinations.

NUCLEAR SAFETY

Containment Systems Experiment

An initial large-scale aerosol transport shakedown run was successfully performed in the CSE drywell to evaluate the adequacy of equipment and techniques for use in such tests in the CSE containment vessel. In addition, the run provided mass transport data on the behavior of iodine and cesium in a steam and air atmosphere for comparison with predictions of a mathematical model developed from the smaller scale Aerosol Development Facility (ADF) data. Gas-Phase concentration of the iodine and cesium aerosol decreased

exponentially with an 8-minute half life compared with a 12-minute half life predicted. Data on gas velocity was obtained, and modification of the mathematical model to utilize the measured gas velocity is expected to result in closer agreement between calculated and observed initial half lives.

A blowdown run was performed with cold water expelled by gas at an initial pressure of 1550 psig. About 100 ft³ of water was discharged in three seconds. This run completed the group of four cold water blowdowns. The performance of instrumentation for liquid weight, liquid level, vessel and nozzle pressures and vessel reaction force has been shown to be adequate in these runs for the transient inputs expected in forthcoming hot blowdowns.

Radioactive Waste Solidification

The third spray solidification run in WSEP was postponed by failure of the melter furnace. Cause of the failure is unknown.

Pots filled in WSEP with radioactive solids showed no measurable change in diameter.

The change in heat within a pot of solid caused by the escape of gamma energy is insignificant. Essentially 98% of the heat is generated inside of the pot and must be transferred to the pot surface.

When sodium (as sodium nitrate) is added to PW-1 waste solution, the viscosity of the evaporated solution at B.P. 140 and 150 °C is significantly reduced. However, with increasing sodium addition there is an increasing formation of a solid component which settles out fairly rapidly at the boiling point.

A 24-hour in-pot melting run was carried out successfully on a PW-2 flowsheet in a nonradioactive test.

Fission Product Aerosol Containment

Depletion of airborne methyl iodide in a steam atmosphere by 5% hydrazine-5% ammonium hydroxide water solution directed down the walls of the chamber occurred when the hydrazine flow rate was made equal to the spray rate used in earlier runs. The rather minor dependence on factor other than liquid throughput for a given hydrazine concentration gives further evidence that the reaction rate in the liquid may be limiting the removal rate.

An equation was derived to fit experimental data obtained for the partition coefficient for methyl iodide between air and water. No effect of concentration was found from 6.4×10^{-9} g mole/liter to 1.28×10^{-7} g mole/liter.

Geophysical Exploration of Rattlesnake No. 1 Well

In-well investigations were completed and the well replugged on June 12, 1967. Interpretation of the large amount of geologic and hydrologic data collected is in progress.

Disposal of Reactor Off-Gas into Soil Systems

Calculations were completed for obtaining minimum gas travel time to the ground surface as function of well depth, injection section length, and injection pressure.

Columbia River Sedimentation Studies

Radionuclides measured in the interstitial water of tidal flats in the Columbia River estuary appeared to originate more from fallout than from reactor effluent. Radionuclides in bottom sediment of the estuary appeared in highest concentrations on fractions with large surface area, as expected.

Simulation Modeling of Thermal Generation

Thermographs were installed on the Illinois River and other steps taken to provide field verification of both the Illinois and Deerfield River cases in order to provide convincing confirmation of the digital simulation model.

Pressure Vessel Crack Monitoring

Development work continued on a program to apply acoustic emission methods to monitor flaw movement in reactor pressure piping. Comparison of data from recent tests using small pressure vessels, formed from 3" diameter pipe, with that from earlier tests indicates that an oxidized surface layer contributes significantly to the detected emission. It appears that as a deformation dislocation encounters the sample surface, the brittle oxide layer effectively amplifies the signal by adding its own emission to the original signal from the dislocation.

Several tapes of acoustic emission data were taken to the Naval Ordnance Laboratory in Corona, California, for processing through their large digital analyzer. Considerable insight was gained as to the spectral content of these signals which gives additional confidence that a usable signal analyzer for an acoustic emission monitor system can be developed.

Two, mult layer, electrostatic, transducers have been locally fabricated for laboratory evaluation, and photo-resistive techniques are being investigated as a possible method for obtaining improved alignment of mask assemblies. Additional work is in progress to produce a prototypic, high temperature, piezoelectric transducer for laboratory evaluations. This type sensor is directed toward the ultimate use of a high temperature piezoelectric such as lithium niobate.

DIVISION OF REACTOR DEVELOPMENT AND TECHNOLOGY PROGRAMS F. W. Albaugh)

CIVILIAN POWER REACTORSNuclear Systems and Concepts Analysis (E. A. Eschbach)Systems Analysis Task Force Activities

Redefinition of fuel cost areas, revised cost area electrical demands, revised fossil fuel costs, and capital costs have been completed.

Computer production runs and debugging runs of linear programming optimization systems have been made for concluding Phase II and beginning Phase III of the Task Force requirements. Some delay in progress has been due to the regrouping of PACTOLUS data, thus proliferating minor problems in LP solutions and report generation.

Debug Data - Phase II

The final "Phase II" 30-year burnup histories for 17 variations of the fuel enrichment in a light water moderated reactor, and five variations of sodium cooled fast reactors were completed. The significant difference from the previous calculation was the use of a constant 80% load factor so that the PACTOLUS code could provide histories at higher or lower load factors for base load or peaking plants from the same burnup data. The cross section data were also slightly revised based on calibrations conducted in the past five months.

D20 Fuel Cost Study

In a continuation of the study on fuel costs in the D20 lattice like the PRTR, credits of \$10/g for U-236, \$50/g for Np-237, and \$150/g for Pu-238 were applied to the fuel costs for thorium systems enriched with U-235. Results of this showed a decrease in the minimized fuel cycle costs from 1.14 to 0.930 mills/kWh_e for the thorium metal driven at 60 W/g at a fabrication rate of \$40/kg. Included in this study were the effects of variations in fuel density and assembly pitch on minimized fuel costs. Results of this portion of the study indicated only a small reduction (~0.6%) in fuel costs if the pitch is increased to 9 inches and no change in fuel costs if the density is reduced from 100% to 70% of theoretical density. This latter result could encourage development of longer lifetime fuels at reduced densities.

Code Development

A literature search through the indices of Nuclear Science Abstracts was made seeking thermal scattering cross section data for H_2O and D_2O later than 1960. Specifically, temperature-dependent formulae were sought which might serve to update those used in the WESCOT subroutine of JASON. The search did not indicate the existence of any more recent formulae than we are presently using.

All versions of the fuel cycle analysis chain have been converted to the 1108 and with the exception of an undetectable problem in the recent four-group ALTHAEA are functioning normally. The execution time on the 1108 is about four times faster than observed on the 1107, but turnaround time is much slower than observed before the 1107-1108 conversion.

Due to recent improvements in the JASON code, it now prints out more of the input data than previously and more output is available for ALTHAEA. An improvement was also made to the PLOTTER link of the chain during the conversion.

Conceptual Reactor Design Studies (J. C. Fox)

Thermal Sink Limitation Study

Gross heat capacity (i.e., the amount of heat required to raise the temperature of the stream 1 °F) calculations have been completed for the major streams in the Lower North Atlantic Slope Basins, New York to York River.

Streams studied in this area are the Delaware, Hudson, Mohawk, Potomac, and Susquehanna Rivers.

Steam-Cooled Fast Reactor

The table of results presented last month has been revised somewhat as a consequence of revised initial reactivity requirements. The new initial reactivities came about from a more detailed calculation with the point burnup code. Inclusion of Pu-241 and Pu-242 has in effect lowered the initial reactivities required for 50,000 MWd/tonne exposure. The revised tabulation was used as a hand-out at the Alternate Coolant Task Force meeting in Oak Ridge, Tennessee, on June 22, 1967.

In order to determine the effect of varying pin size, calculations were made for different diameters using ORNL cross sections, from HRG and TEMPEST. Increasing the pin diameter causes slight increases in k_{eff} and slight decreases in both core and blanket breeding ratios. The increase in k_{eff} can be used to decrease

enrichment with an accompanying increase in breeding ratio, sufficient to make the previously mentioned decrease negligible even in the wettest cases where it is largest. It is the thermal hydraulic and economic analyses that will determine the pin diameter between 0.15 and 0.40 inch rather than reactor physics considerations.

Doppler coefficients were obtained from FCC by varying the temperatures for each isotope which must be specified with the Russian cross sections. About all that can be said about the results are that they seem to be the right order of magnitude, appear to be due to Pu-239, U-238, and to a lesser extent Pu-240, and that the trends between cases may be shown to be real by better calculations.

Deuterium was substituted for hydrogen in FCC calculations with the Russian cross sections. Both the reactivities and core breeding ratios are higher for heavy steam. The increase in core breeding ratio is a function of the coolant inventory, lbs steam/ft³ of core, as might be expected. It is about 0.03 for A cases, 0.12 for B cases, and 0.20 for C cases. The increase in reactivity will mean a more negative void coefficient and further increases in breeding ratio due to a lower enrichment. This is particularly true for case 1C.

A comparison of the results of HRG with ORNL cross sections and FCC with Russian cross sections was carried out. The breeding ratios for the latter are consistently higher. This was mentioned by Hannerz, Ekholm, and Von Bonsdorf at the San Francisco meeting last April. Furthermore, the enrichments vary in such a manner as to lead one to the belief that hydrogen cross sections are also unrealistic in one of the cross section sets. We have managed to run ORNL cross sections in FCC. In the one case which allows comparison, OA, due to the lack of necessity for a thermal group, the k_{eff} are 1.0160 and 1.0151 for ORNL and Russian cross sections, respectively. The core breeding ratios are 1.2249 and 1.3328. More detailed comparisons will be necessary. Burnup and shape factors will be rerun with ORNL cross sections in order to be consistent.

The results of the PNL parametric physics study have been incorporated into the design and economic simulations of the SCFBR. The enrichment, and over-all breeding ratio, as functions of core buckling, volume fraction fuel and steam inventory (lbs/ft³ of core) are used in the simulations. From the enrichment the fissile inventory and thru-put are calculated for use in the fuel cycle cost calculations. The specific inventory (Kgs/MWe) of fissile material and the doubling time (years) are estimated for each specific design to show its performance as a breeder reactor.

The incorporation of the physics information made the design and economics simulations complete enough to begin meaningful parametric studies. Since the PNL study is to augment the ORNL evaluation of two specific SCFBR's, the design and operating parameters of one of these designs (Karlsruhe, D-1) was used to check the

simulation. A preliminary parametric study was also made around this design. The results of this study were presented to the Alternate Coolant Task Force meeting at ORNL on June 21 and 22.

Several adjustments in the design simulation were required to get the performance estimated by Karlsruhe for the D-1 design.

The clad temperature calculated by the simulation was about 150 °F higher than Karlsruhe predicted. Part of this difference was traced to a 15-20% higher heat transfer coefficient used in their calculations. Karlsruhe referenced experimental work with rod bundles which indicated this difference in heat transfer coefficients. The basic correlations of Sutherland and Westinghouse are developed from data obtained in heated annuli. The increased heat transfer coefficient is now used in the PNL simulation. The hot channel factors were also modified to match the Karlsruhe analysis.

There is a significant difference in the pressure drop of the D-1 core as calculated by the PNL simulation and Karlsruhe (60 psi vs 136 psi). The cause of this discrepancy is still being investigated.

The physics information in the simulation, enrichment and breeding ratio, will be reasonably close to that predicted by Karlsruhe and ORNL for the D-1 design when the latest results are incorporated.

The parametric study performed on the D-1 design covered a system pressure from 1800 psia to 3000 psia. At 1800 psia the pumping power requirements of the D-1 design limited its feasibility and lower pressures were not investigated. A sensitivity study will be carried out around this 1800 psia point to determine what changes can be made to the D-1 design (core volume, height to diameter ratio and fuel pin size) to allow operating it at lower system pressures. The incentive to go to lower pressures is to increase the breeding ratio.

The PNL design simulation will also be modified to perform a parametric study of the B&W 1250 psia SCFBR design. The simulation must be modified to do this because the B&W design is very compact. This results in reduced pumping power requirements due to lower out-of-core pressure drop. It represents a major difference in design philosophy from the Karlsruhe D-1 design. By considering these two design philosophies, the PNL parametric study results should span other design concepts that could be proposed.

The clad design analysis that was proposed for use in the PNL design simulation was found to have a limited range of applicability. To cover a wider range of pin diameters, clad thickness and system pressures, additional modifications will be made.

The fuel cycle cost simulation has been debugged; however, more consistent unit costs and process times must be established with ORNL before final studies are performed.

The analytical model predicting blower efficiency in the design simulation has been abandoned because of the different blowers proposed for use in steam-cooled fast reactors. When considering the D-1 design, a blower efficiency of 76% will be used. When considering the B&W design, a value of 85% will be used. The D-1 design has a multistage axial flow compressor while the B&W design has a single stage radial flow blower. The constant efficiency model will be less accurate at higher blower powers than the previous model, but this should not distort the parametric study results.

USAEC-AECL Cooperative Program (J. J. Cadwell)

Cross-Flow Mixing Between Parallel Flow Channels During Boiling

Analytical studies and laboratory experiments to determine values of cross-channel mixing during two-phase flow in rod bundle fuel elements were completed. The basic purpose of this program was to obtain a method to predict flow and enthalpy in each subchannel of rod bundle fuel elements as influenced by cross-flow mixing. This mixing is important in the analysis of fuel element performance and in the determination of possible boiling burnout conditions for bundle-type fuel elements.

The results of the analytical phase of this study is described in the report "Cross-Flow Mixing Between Parallel Flow Channels During Boiling - Part I - COBRA - Computer Program for Coolant Boiling in Rod Arrays" (BNWL-371 Pt 1), which was issued earlier. The analytical model used in COBRA includes the effects of two types of cross flow mixing between the subchannels. The first type is diversion cross flow caused by unequal pressure distributions in the adjacent interconnected channels such as would result from different boiling lengths in each of these channels. The second type is turbulent cross flow caused by the random travel of coolant between adjacent subchannels. The first type of cross flow can be computed fairly well from the equations of the mathematical model; however, the turbulent cross flow cannot, and must be determined from experiments.

In the experimental phase of this program, two sets of experiments were performed to supply values of the turbulent cross flow in "clean" subchannels (no warts, wire wraps, etc.). In the first set of experiments an electrically heated test section which simulated two adjacent subchannels formed by rods on a square array located adjacent to rods on a triangular array was used. Measurements were

made of subchannel flow rates and exit enthalpies, and this information was used to determine cross-flow mixing during boiling by comparing the results of the experiments to calculations using COBRA. The results showed that mixing during boiling was doubled over the nonboiling case when the simulated rod spacing was 0.084-inch; however, for a spacing of 0.020 inch, no significant improvement was observed. A report describing and analyzing these results is under preparation and will be issued as BNWL-371 Pt 2.

For the second set of experiments an electrically heated test section which simulated two adjacent flow channels formed by rods on a square array was used. Because the two channels had equal flow areas and heat inputs, the only significant mixing that occurred was the result of turbulent mixing through the gap between the two channels. The amount of mixing that occurred over the length of the test section was measured by injecting tracers into the inlet stream of one of the two parallel channels and measuring the tracer concentrations at the exits of the channels. Detailed analysis of the results is presently under way. A report for these experiments will be issued as BNWL-371 Pt 3.

Significant conclusions resulting from both of these experiments are as follows:

1. Mixing is not proportional to rod spacing. Both experiments showed that, during nonboiling conditions, the natural turbulent cross flow per unit length is nearly independent of spacing and can be correlated as a function of Reynolds Number. Successful correlation of these data is significant because it is the lower limit of mixing in actual rod bundles.
2. Mixing during boiling was found to improve from 0 to 300% over the nonboiling case depending on test section heat flux, flow rate, pressure, and rod spacing.
3. Reducing pressure from 750 to 400 psia caused increased mixing during boiling at a given flow and heat flux.
4. A reduction in flow rate caused increased mixing during boiling at a given heat flux and pressure.
5. If mixing is assumed to be a function of quality for a given flow rate and pressure, a peak in mixing occurs at about 10 to 20% quality.
6. At 0.020-inch spacing very little improvement in mixing occurs during boiling; however, the same effects noted above apply.

7. The relative heavy water and lithium ion tracer concentrations at the test section exit were nearly the same indicating that both the vapor and liquid phases are active in the mixing process during boiling.

The results of the experiments have demonstrated the validity of the analytical methods used in COBRA, and they have provided information vital to the application of COBRA to subchannel analysis. The experiments have also answered questions concerning the nature of mixing during boiling and have identified some of the significant parameters involved.

Effects of Rod Spacing on Boiling Burnout

Construction of an electrically heated test section to study the effects of rod spacing on boiling burnout in multirod bundle fuel elements is nearly complete. The test section will be used to investigate the effect of adjacent heated rods on boiling burnout by using a single rod surrounded by four simulated rods. The simulated outer rods are formed by a 4-lobe tube. The four convex surfaces facing inward simulate the adjacent heated surfaces, and the concave inner surfaces form the flow subchannels. The wall thickness of the concave inner surfaces is reduced at the outside to provide the proper heat generation in the vicinity of the subchannels. Design of the test section allows different diameter inner rods to be inserted, thus allowing adjustment of the rod spacing. Several inner rods will be fabricated to allow spacings from 0.015 to 0.080 inch. The test section can be operated with either the inner or outer surfaces heated or with both surfaces heated. Laboratory experiments with this test section are scheduled for FY-68.

Evaluation of Zr-2.5 wt% Nb Pressure Tubing

The general objective of this program is to evaluate Zr-2.5 wt% Nb alloy tubing as a pressure tube material with reference to tests and reactor experience that has been obtained on Zr-2 pressure tubes.

Four fatigue tests have been completed on heat treated and aged tubular test specimens with electrical discharge machined (EDM) slots cut into the outer surface. The finished EDM slots were 10 mils or less in width and semicircular in profile with radii of 0.125, 0.100, 0.075, and 0.050 inch. The peak hoop stress for each of the four test specimens was about 23,000 psi, and the cycling rate was about 400 per hour. In each test, leakage of the internally pressurized water occurred at a fatigue crack length of about 0.41 inch, irrespective of the initial EDM slot size. This shows that leakage is a function of tube geometry and internal pressure.

These four tests were continued until failure occurred. The fatigue crack lengths at failure were 1.36, 1.31, 1.08, and 0.95 inch for the 0.125, 0.100, 0.075, and 0.050 inch EDM slot radii,

respectively. The number of pressure cycles to produce fatigue failure were 18,600, 25,000, 35,500, and 47,200 for the above EDM slot size order. These test results suggest that the critical crack size may be partially dependent on the extent of prior fatigue damage and that the critical crack size decreases with increasing amounts of prior fatigue damage. Additional tests are planned to further substantiate these observations.

Hydraulic and Nuclear Stability in Parallel Flow Channel Systems

The analog computer study of hydraulic and nuclear stability of the BLW-250 has been completed. Final results have been presented to the AEC-AECL Technical Advisory Committee (TAC) and the Subcommittee on Reactor Stability and Control. A rough draft of the final report was distributed to the members of the Subcommittee, and comments have been received. These comments are currently being incorporated into the final report.

In-Reactor Measurements of Creep in Zr-2.5 Nb Alloy

An in-reactor stress rupture test was conducted at 400 °C and 38.7 kg/mm² (55,000 psi) stress. The specimen ruptured in 156 hrs with 23% total elongation and 61% reduction of area. This ductility is in good agreement with values of 21% reported in the March monthly report and with the total elongation of a control creep test and a tensile test conducted on an in-reactor pre-crept specimen, both at 400 °C.

In-Reactor Corrosion of Zirconium Alloys

Zirconium alloy corrosion specimens exposed in the G-7 loop of the Engineering Test Reactor are undergoing postirradiation examination at Battelle-Northwest. The specimens were exposed in pH-10 NH₄OH, <0.05 ppm O₂ at 270-280 °C. Specimens from the following holders are being examined.

8-Cycle Exposure (174 days):

Quadrant 235 exposed at $\sim 1 \times 10^{14}$ n/cm² sec, >1 MeV*
 Quadrant 233 exposed at $\sim 1 \times 10^{13}$ n/cm² sec, >1 MeV*
 Out-of-flux holder ~ 1 sec from the flux
 Out-of-flux holder ~ 1 min from the flux

4-Cycle Exposure (84 days):

Quadrant 243 exposed at $\sim 1 \times 10^{14}$ n/cm² sec, >1 MeV*

*Estimated Fluxes; flux monitors not yet processed.

Initial oxide weight gains were obtained but will not be reported until chemical cleaning and weight checks are completed. Hydrogen

samples, taken from selected specimens from all of the holders, are undergoing analysis.

A black deposit was present in various degrees on specimens from all of the holders. On specimens from the out-of-flux position ~1 min from the flux, the deposit was thin. At the low flux position (Quadrant 233), the deposit was relatively uniform on all specimens. At the high flux position (Quadrant 234), the black deposit occurred only on a fraction of the surface of most specimens, appearing to have spalled from a large fraction of most of the surfaces. Quantitative values at the high flux position (Quadrant 234) therefore have little meaning in terms of uniform crud deposition. The heaviest deposit occurred at the out-of-flux position ~1 sec from the flux. The loop stream undergoes an abrupt change of direction near the holder, which likely contributed to the heavy deposit.

Since the deposit on Quadrant 233 specimens was relatively uniform, crud deposit weights on each specimen were determined to compare deposition as a function of alloy and surface pretreatment (etched versus prefilmed). Crud weights on 8-cycle specimens are reported in the table.

Crud deposition data for previous exposures in the series were summarized in the January 1967 monthly report. However, comparisons with the data in the table are complicated by a loop decontamination during the 8-cycle exposure. The specimens were removed from the loop during the decontamination, but the loop crud inventory changed during the 8-cycle exposure. In this report only internal comparisons of 8-cycle data will be considered. Summarizing the observations:

- As a function of loop position, deposition was low immediately above the flux zone (~1 min position), slightly higher in the flux zone, and very much higher immediately below the flux zone (~1 sec position), at a point where the loop stream changes direction.
- There was no consistent effect of initial surface condition on crud deposition.
- There are no marked deposition trends as a function of alloy; however, deposition was slightly lower on specimens containing niobium.
- There was no consistent pattern of crud deposition as a function of sample position within the specimen holder.

CRUD DEPOSITION ON SIX ZIRCONIUM ALLOYS AFTER 8-CYCLES (174 Days)

Deposition at In-Flux and Out-of-Flux Positions

	Maximum Deposit mg/dm ²	Minimum Deposit mg/dm ²	Mean Value mg/dm ²
~1 Min from flux	9	2	4
Quadrant 234	19	2	6
Quadrant 233	12	3	7
~1 Sec from flux	351	56	210

Deposition in Quadrant 233 as a Function of Alloy

Alloy*	Surface Condition	
	Etched mg/dm ²	Prefilmed mg/dm ²
Zircaloy-2	7.0	7.4
Zircaloy-4	10.2	6.7
Zr-1.2Cr-0.08Fe	11.0	9.9
Zr-1.2Cu-0.28Fe	10.1	4.8
Zr-3Nb-1Sn	3.5	11.9
Zr-2.5Nb (Q 20 CW A)**	5.1	4.8
Zr-2.5Nb (Q 30 CW A)**	5.7	8.8
Zr-2.5 Nb (Ann 20 CW)**	4.9	5.2

*Mean weight gains on duplicate or triplicate specimens.

**Q = quenched; CW = cold work; A = aged; Ann = annealed.

HTGR Graphite (R. E. Nightingale)

Graphite-Water Vapor Reaction Under Gamma Irradiation

The experimental apparatus previously employed to investigate the reaction of graphite with water vapor under gamma irradiation (see HW-83737) was extensively modified. The purpose of the modifications were threefold: (1) To remove those portions of the system where oxygen could be trapped when the system was open and then bleed into the gas stream during subsequent operation. (2) To remove metals which could be oxidized from contact with the gas stream, particularly in high dose rate - high temperature regions. These metals included heater windings, power leads, thermocouple leads, and a gold chain used to suspend the sample. (3) To remove standard-taper joints which, because of the gamma radiation and/or high temperature, could not be greased or waxed and hence were potential sources of gas leakage.

Following return of the modified system to operation, three runs were performed. A sample of TSX graphite weighing approximately 8.2 grams was exposed to a helium stream containing about 110 vpm water vapor and flowing at 200 cm³/min. The gamma dose rate at the sample position is about 2×10^7 r/hr. The oxidation rates measured at the three sample temperatures employed are tabulated below:

<u>Sample Temp. (°C)</u>	<u>Oxidation Rate (hr⁻¹) $\times 10^7$</u>
536	1.56
633	2.65
694	9.55

The highest oxidation rate observed is about the same as the lowest rate, previously measured under similar reaction conditions, with the exception that the dose rate at that time was 2.86×10^6 r/hr. This indicates that the revisions to the system largely eliminated oxygen from the gas stream.

Oxidation of TSX Graphite by Water Vapor

A preliminary electron microscope study was made of the oxidation of TSX graphite by water vapor. The transmission microscopy specimen was cut from graphite rod stock using a high speed alundum cutoff wheel. When the specimen was subjected to 15.4 mm Hg of water vapor in a helium stream at 960 °C for a period of 18 hours, a 3% reduction in specimen weight resulted. Microscopic examination indicated that a light selective oxidation had occurred which was qualitatively similar to the thermal reaction between TSX and oxygen at 700-800 °C. The selective attack exposed graphite layer plane structures, and there was considerable evidence of pitting of the layers involving holes of various sizes, some with hexagonal shapes. There was also some graphite hillock formation.

Dragon Water Injection Test

During August 18 and 19, 1966, a water injection experiment was conducted in the DRAGON Reactor. This experiment consisted of injecting water into the circulating helium coolant over a period of 12½ hours. The quantities injected during each half-hour period were reported, but it is not clear whether the water was added continuously or by rapid injection at the start of each half hour. Gas analyses of H₂O, CO, H₂, CO₂, and CH₄ were made, but the analysis accounts for only about 50% of the water injected. However, knowledge of the relative quantities of the gases is useful in interpreting the results. Maximum quantities of gases observed were: H₂O 6 vpm, CO 3 vpm, CO₂ 1 vpm, H₂ 8 vpm, and CH₄ 0.7 vpm.

The water injection test was simulated using the GOP computer program and the pertinent geometry, flow rate, and temperature

distribution in the DRAGON Reactor. The calculated results show that radiation reactions are required to obtain as much CO_2 as was observed. Without the radiation reactions the CO/CO_2 ratio would have been about 100 and the CO level would be nearly equivalent to the hydrogen level.

Either method of injection, i.e., continuously or rapid, can be simulated on the computer. The two methods result in different levels of the gas composition being obtained at the end of any injection period. During the final seven hours of the test, each half-hour period had about the same "injection rate." If we assume that the water was injected during this period in a "constant leak rate method," it is possible to calculate the steady state $\text{H}_2/\text{H}_2\text{O}$ ratio for the DRAGON conditions. The calculated value was 1.09 which agrees well with a value of 1.2 determined from the gas analysis during this period. This agreement may be fortuitous, however, because of the lack of material balance in the gas analysis and the uncertainty in the injection method. The calculated H_2 and H_2O levels were 11.5 and 10.6 ppm compared to the approximate peak analysis values of 7.5 and 6.35 ppm, respectively.

APPLIED AND REACTOR PHYSICS

Plutonium Criticality Studies (E. D. Clayton)

Pulsed Neutron Source and Reactor Noise Experiments

A series of pulsed neutron source experiments have been performed on heterogeneous plutonium systems made up of layers of PuO_2 -polystyrene and Lucite. The base dimensions of the core (a rectangular parallelepiped) were 36.3 cm by 31.0 cm. The assembly was reflected on two sides (ends) by one inch of Lucite. The core was comprised of four two-inch slabs of PuO_2 -polystyrene material (2.2 wt% Pu-240) at an H/Pu of 15, with two inches of Lucite between each fuel slab.

The reactivity was measured as a function of assembly height during the approach-to-criticality. The estimated height of delayed criticality was 34.9 cm. The data were analyzed by means of the Garellis-Russell method, assuming an effective value for the delayed neutron fraction of 0.0025. The results are summarized in the following table. The measurements were performed with a Kaman-Nuclear A-808 neutron generator. However, during the course of experiments the neutron yield decreased significantly, which precluded measurements with the system further subcritical (-4.5 dollars). The causes for the decreased neutron output of the generator have been determined and are being corrected for in a replacement tube.

Pulsed Neutron Source Experiments

<u>Assembly Height (cm)</u>	<u>Reactivity (Dollars)</u>	<u>Effective Reproduction Factor</u>	<u>Decay Constant α (sec⁻¹)</u>
20.3	-4.47	0.9889	4494
25.4	-3.05	0.9924	3152
30.5	-2.64	0.9934	1389
31.2	-2.99	0.9926	1179
31.8	-2.09	0.9948	930.2
32.5	-1.46	0.9964	756.6
33.0	-1.56	0.9961	550.2
33.8	-1.15	0.9971	372.9

A Rossi alpha type measurement of the neutron decay constant was attempted on an undermoderated plutonium assembly with poor results. The conditional probabilities for detection of a second neutron relating to the same chain as the neutron initially detected were essentially the same. It is thought that inadequate discrimination between the gamma rays and neutron pulses originating in the boron loaded liquid scintillator may be the cause. Work is now proceeding on pulse shape discrimination equipment for separation of the neutron and gamma pulses.

Minimum Critical U-235 Enrichment for Uranium Nitrate Solutions

The value of the minimum critical enrichment (the smallest uranium enrichment that can be made critical) for a homogeneous mixture of slightly enriched uranium is of considerable importance for nuclear criticality safety in the fabrication, shipment, and processing of slightly enriched reactor fuels. Knowledge of the enrichment also provides a clean simple case ($k_{\infty} = 1$) for checking criticality calculations for slightly enriched uranium.

Experiments previously completed in the PCTR have determined the limiting critical enrichment for homogeneous hydrogen moderated mixtures of uranyl nitrate, $\text{UO}_2(\text{NO}_3)_2$, to be 2.1 wt%.* Ninety-nine percent confidence limits have now been established at ± 0.03 wt%. The value computed by means of the GAMTEC-II code is 2.09 wt%, which agrees well with the experiment.

Based on these results the single parameter limit for nuclear criticality safety of uniform aqueous solutions will be 2.07 wt% for uranyl nitrate solutions. This value is subject to the condition that at least two nitrate ions will be present for each uranium atom. The limiting critical enrichment of 2.07 wt% has now been

*Experiment partially funded by National Lead Company of Ohio.

included in the proposed revised U.S.A. standard, Nuclear Criticality Safety Standard for Operations With Fissionable Materials Outside Reactors, prepared by Subcommittee ANS-8 of the American Nuclear Society.

Phoenix Fuel Reactor Program (D. D. Lanning)

CAF-Phoenix Fuel Experiments

Preliminary analysis of the noise data from the CAF experiments has been completed. The values obtained for $\frac{\beta-\rho}{\lambda}$ appear to be good with fractional standard deviations of approximately 2%. A few more computations will be necessary to extrapolate the values of $\frac{\beta-\rho}{\lambda}$ to $\rho=0$ for the final $\frac{\beta}{\lambda}$ result. However, an estimate of these are 630 for $\frac{\beta}{\lambda}$ and 50" for the critical height of an array of 19 fuel columns λ with the center column replaced by a poison column. Radially, the reflector contains 2 inches of beryllium in addition to water.

CAF-Phoenix Fuel Calculations

The three experimental approach configurations have been encapsulated in a PHYSICS CHAIN input set. This input provides a means of evaluating new cross sections according to the CAF experiments. The cross sections must be contained in a PHYSICS CHAIN library tape. The cross sections are spectrum-averaged, inserted into the diffusion theory model used in the analysis of the CAF and k_{eff} is calculated for the three extrapolated-to-critical assemblies. Thus, the ability of a cross section set to predict the correct multiplication for a hard spectrum ($\rho_{FAST}/\rho_{THERMAL} \approx 6$), strongly reflected plutonium-fueled system can be checked using seventeen group diffusion theory. Other energy groupings may be evaluated by a minor input change.

This set of rather clean but realistic experiments gives a "bench mark" for future calculational methods and cross section sets.

PRCF-Phoenix Fuel Experiments

Analysis has been completed of the power distributions measured in the 3x9 shimmed core. The measurements included horizontal traverses across the plates of fixed elements and axial traverses along the plates of a fuel follower of a shim which was 59% withdrawn. The axial traverse of the shim fuel follower shows that the axial peak power on an outside plate is located at 0.1" below the bottom of the meat in the fixed fuel element. The magnitude of this peak was found to be no greater than the peak observed in an exterior plate of the fixed element. The horizontal traverse obtained from the plates of the fixed elements show the characteristic power peaking at the edge towards the beryllium reflector.

Irradiations were also made with plates containing tapered fuel cores of $1\frac{1}{2}$ " and $\frac{1}{2}$ ", respectively. Results of the gamma scanning measurements indicate a reduction of at least 30% in the power peaking at the bottom edge of the tapered plates as compared to a standard fixed fuel plate.

An additional irradiation was performed on plates sectionalized into 3 - 9 - 3 inch wafers in support of a Phoenix fuel irradiation test scheduled at MTR in the near future. Gamma scans were made, and analysis of the axial traverse along the plates is in progress.

Measurements were performed to determine the worth of the mockup regulating rod in the PRCF. Full-out to full-in gave a reactivity change of -7.4% , and full-in to 11 inches out gave a reactivity change of $+4.5\%$. Additional analysis will be required to translate these results into the worth of the regulating rod in its normal position in the MTR.

Temperature measurements were performed as part of the exploratory measurements, which were initiated to isolate the cause of the slow loss of reactivity observed in the operation of this core. These measurements were found to be extremely sensitive to the time that the H_2O was left in the reactor tank following the periods of heating. This was believed to be due to cooling on the uninsulated top and bottom reflector surfaces. Measurements made immediately following moderator mixing displayed consistent results as shown by moderator temperature vs reactivity curves. The temperature coefficient obtained from this data indicates a value of $\sim -1.9\%/^{\circ}C$ over an operating range of 31 to 40 $^{\circ}C$.

An additional problem associated with this core is the probability of a source of neutrons from the (γ, n) reaction in the beryllium reflector. To decrease the relative sensitivity of this effect on the operation of this core, several of the chambers were relocated, and process specifications were changed to allow operation at ~ 1 kw instead of the normal 100 watts. This additional decade increase in operating power allows adequate waiting to reach time asymptotic periods and permits measurements with shorter periods.

PRCF-Phoenix Fuel Calculations

The analysis of the PRCF-Phoenix experiments has started. An examination of the core composition data has shown a comforting uniformity of weight percent plutonium and isotopic composition. The isotopic analysis of alloy heats has been weighted according to the number of plates used in the actual core.

Initial effort has been directed towards a model for the simplest, most symmetrical unrodded critical core of 14.25 fuel elements. This experiment is amenable to two dimensional analysis and will best lend itself to establishing the mesh and group

sensitivities in the most economical and rigorous manner. Since the later full core rodged experiments will require a full three dimensional analysis with only two energy groups (a code limitation), the effect on k_{eff} of collapsing to two groups from some much larger number is of great interest. This pure core experiment will allow reasonably rigorous treatment of this effect.

MTR-Phoenix Fuel Calculations

Recent data available from the PRCF Phoenix experiment has provided a means to check the shapes of the calculated power distribution in the MTR Phoenix core. Selected fuel plates in both a stationary and shim fuel element were gamma scanned following irradiation. The gamma activity is proportional to the power generated and thus gives the relative power generated in the plate as a function of axial position.

By normalizing the area under the measured power distribution to equal the area under the calculated power distribution, it was possible to compare the magnitudes of the power peaks and the overall shape of the power distribution. In general, the measured and calculated power distributions have the same shape over the length of the fuel plate. The exception to this general agreement was along the bottom water reflector-fuel interface. At this position, the magnitude of the measured power peak was approximately 30% to 50% smaller than the calculated values.

The cause of this rather large difference in the magnitude of the power peak is not fully understood at this time and will require further investigation. Lowering the value of the expected power peaks will significantly raise the permissible power level for the MTR.

Code Conversion. The HAMMER system of chained programs was modified to operate on the UNIVAC 1108 and is now operational. The HAMMER program of tape U9387 operates with the same input requirements as the 1107 version. An alternate version of the program exists on tape U0274 which utilizes drum storage for both the library tape and the scratch tapes. The thermal and epi-thermal libraries are on the program tape and are assigned to drum files at execution time. Further information on the operation of this version of the HAMMER program can be obtained from W. W. Porath, 3702 Building, 300 Area, phone 3774.

MTR Core - Thermal Hydraulic Analysis

A preliminary thermal hydraulics analysis of a Phoenix core in the MTR has been completed. Power profiles were taken from calculations made by Engineering Physics personnel with some modification based on results obtained from the PRCF experiment with the Phoenix core. Limitations assumed on heat fluxes and incipient nucleate

boiling require reactor operation at 29MW_t . At this power level the thermal hydraulic conditions in the core are comparable to the present MTR core conditions at 40MW_t operation. Results of this analysis and a detailed description of the methods used were presented to MTR personnel. General acceptance of the methods was expressed.

MTR Core - Mechanical Design

A trip was made to Idaho and the proposed flux monitor system was reviewed with several contacts at INC. The location of the wand down the inside corner of the fuel box appeared satisfactory to those who reviewed it. Most of the comments concerned shaping the flux wand head to allow the use of a simple handling device for removal and insertion. A print was obtained by MTR Operations on a flux wand handling device which they now use to position and remove their flux wands. The information on this handling device will be used to design a handling device for the Phoenix experiment flux monitor wands.

The condition of shim rod No. 42 was further reviewed with MTR Operations. A recent photo was found which showed the lower bearing was still in place. The condition of this bearing and the shock absorber is still speculative. An attempt will be made by MTR Operations people to determine more about the bearing during the next outage. It was thought that possibly a small underwater camera could be lowered down into the reactor and with adequate lighting view the bearing and shock.

Two mockup MTR Phoenix fuel boxes are being fabricated by Nuclear Metals Division of National Lead and are scheduled for completion by mid-August 1967. These mockup fuel boxes contain 16 simulated dummy fuel plates and have the provisions to accommodate a flux monitor wand. The flux wands are being fabricated at BNW; however, they will not have a fueled core.

The mockup fuel boxes will be used to evaluate the proposed flux monitor well and to develop a satisfactory handling device for removing and reinserting the wands. The boxes will also be hydraulically tested at Idaho in the hydraulic test facility at 140% of rated MTR flow.

MTR-Phoenix Experiment

An irradiation experiment is being prepared to test plutonium fuel plates under the operating conditions proposed for the MTR Phoenix experiment. The purpose of the test is to demonstrate in-reactor corrosion resistance as a function of exposure in the Phoenix fuel geometry. The first plutonium fuel plates prepared for the irradiation test had internal voids and inclusions associated with the cast cores. These plates were not suitable for the planned

irradiation, and new plates were prepared using extruded Al-Pu core material. These plates had no internal defects within the core and the plutonium contents were exceptionally uniform.

The six fuel plates (three each, 1100 and 6061 cladding) for the long (9") irradiation test section were completed, and the short (3") fuel plates are being fabricated. However, the external hardware, which holds the test sections, and which was previously used for irradiation tests in the MTR, was found to be too radioactive to be used for this test. BNW will fabricate the new hardware from prints furnished by Idaho Nuclear Corporation. Fabrication of the new hardware will delay the irradiation test approximately six weeks.

The 1100 clad fuel plates, in which a 6061 spacer is used to minimize "dog boning," have an unbonded interface between the 6061 insert and the picture frame. The interface is parallel to the rolling direction, and the rolling reductions are apparently insufficient to disrupt the oxide film and cause bonding of the facing surfaces. It is intended to seal this interface by welding. Other courses of action involve developing an alternative design which will permit deformation of the interface or using only 6061 cladding for this test. Preliminary specifications for the fuel element for the MTR experiments have been written. These specifications will be reviewed with all interested parties before the final specifications are prepared.

MTR-Phoenix Experiment Coordination Meeting

A coordination meeting on the MTR-Phoenix Fuel Burnup Experiment was held at Idaho Falls, on June 27, 1967. The present status in the development of fuel specification for full plutonium core loading of the MTR was discussed. Plans for review of these fuel specifications and acceptance of the fabricated fuel were reviewed. Work on the safety analysis and preparation of the MTR core for this fuel burnup experiment will now be initiated by the INC personnel. Information required for the safety analysis report that can be derived from the PRCF mockup experiment is to be obtained before the completion of the critical experiment program.

Applications

HFIR. A meeting was held in San Diego on June 14, 1967, to discuss the possible application of Phoenix fuel in the HFIR. The meeting was attended by interested persons from ORNL and ANL, together with several members of the Phoenix Fuel Program from PNL. Some preliminary comparison calculations on the core reactivity lifetime were discussed. Several questions were raised concerning these preliminary calculations. It was felt that a thermal flux advantage in the flux trap (ITC) should have been found for the plutonium loaded core, especially since a fast flux advantage was

calculated. The extended core reactivity lifetime for Phoenix fuel is of interest. However, fuel development will be required to assure a uniform fuel dispersion and to assure that limitations of fuel lifetime due to radiation damage and surface oxidation are considered. In order to justify any such fuel development, it was felt that a flux advantage should first be shown, or at least not a flux disadvantage. Further study needs to be made to determine if the predicted extension of core lifetime would reduce the operating cost, since there is believed to be a large uncertainty in the additional expense of fabricating plutonium fuel assemblies and development of long life fuel. Calculations on an optimized plutonium fuel loading are to be made to further explore the comparison between the U-235 core neutron fluxes and those obtainable with a plutonium core.

CNSG. Analytical studies of the physics characteristics of Phoenix fueled loadings in the Consolidated Nuclear Steam Generator (CNSG) have continued. Calculations have been performed for PuO₂-ZrO₂ fueled loadings in the CNSG-II core design. The characteristics investigated are:

1. Reactivity variation with changes in enrichment, rod size, and moderator-to-fuel volume ratio.
2. Core lifetime versus enrichment.
3. Temperature coefficients of reactivity (Doppler and moderator).

Core lifetime calculations have also been performed for a base comparison case with 5.3 wt% enriched uranium loading.

A reference thermal hydraulic model of the CNSG-II core has been developed using the REPP computer code for pressurized and boiling water reactors. A complete core design adequate to put directly into the REPP code was not available. The power profile and DNB ratio were adjusted to make the simulation fit the available information as well as possible. A pressure drop value close to the published value for the CNSG-II core could only be obtained by ignoring the effect of spacers. The reference core for the application study will have a higher pressure drop to include the pressure drop effect of spacers. The first physics calculations will then be used to limit the range of interest to that exhibiting the desired Phoenix effect.

High Temperature Reactor Lattice Physics Studies

(R. E. Heineman)

Reactor Construction

The acceptance tests on the horizontal control rod drives were completed during the month, and the units were accepted by the HTLTR Operations and Systems Unit with minor exceptions.

The two experimental oscillators were modified in several respects at the request of the Operations Unit following the successful test of the light-duty oscillator to 1000 °C. The hydraulic drive units and the oscillator controls were installed. These units were accepted by the Operations Unit with the exception that the oscillators be mounted in the reactor room at the reactor faces.

The acceptance tests on the vertical safety rods were interrupted by the inability of the drive motors to pull the rods out of the reactor in a consistent manner. As described later in this report, the gear ratio in one drive unit was modified and the unit reinstalled in the reactor. The subsequent tests at room temperature and at a temperature of about 400 °C were successful. The rod drop time of 0.79 second compares favorably with the 0.75 second theoretical drop time. The vertical rods were accepted by the Operations Unit, subject to completion of several modifications described later.

The drift tube for the neutron time-of-flight spectrometer was installed with a temporary termination at the reactor room. The chopper will be assembled and put into operation only after the nuclear startup experiments have been completed.

Exceptions on some of the equipment previously accepted have been completed. A new solenoid in the gamma monitor made it operational. The high efficiency filters in the filter pit, through which the reactor ventilation air passes, were installed. The temporary flooring in the reactor room and basement was removed. The requisition for a vibration monitor for the main blower was approved. The materials for the limit switches for the VSRs were ordered. The noise on the various analog inputs to the control system was suppressed.

With acceptance of the building, the reactor, and the engineered equipment, the project was officially closed out at the end of the month. Exceptions which will be completed with cost accrual to the project include: installation of the oscillators, completion of modifications to the vertical safety rods, assembly of the neutron chopper, completion of all "As-built" blueprints, and minor items of a finishing or touchup nature.

Reactor Equipment

Oscillators. The fabrication, cold testing, and hot testing of the light-duty oscillator have been completed. Fabrication and cold testing of the heavy-duty oscillator have also been completed.

Modifications to both oscillators have been made as requested by the HTLTR Operations Unit. These include an analog position readout, a handwheel for manual operation, an improved method for lifting the covers, moving the viewing window on the heavy duty unit, making the withdrawal of a 14-foot sample in the light duty unit possible, and removing the hinges and substituting cam lock bolts on the light duty unit.

Valves and hydraulic power supplies have been taken to the reactor building and have been installed. The oscillators and keys will be delivered to the reactor building when the high temperature design test, now in progress, is completed.

Vertical Safety Rods. The vertical safety rods have been installed on the reactor and operated, under computer control. Three problems became apparent during this test operation.

1. The maximum torque available from the rod drives was not adequate to consistently withdraw the rods from the reactor. In the initial design of the rods, the torque available from the drive was designed with a very small margin over that required to withdraw the rod. This was done so that very little excess pull could be put on the rod when withdrawing it; thus, any drag on the rod due to misalignment or other cause would stall the drive leaving the rod free enough to scram back into the core under its own weight. Tests on the drive indicated that it would pull about 75 pounds when operated at design speed (100 pulses per second). The rod weighs 60 pounds, and it has been found that any slight misadjustment of the drive decreases its pull to a point where it does not consistently withdraw the rod. To remedy this marginal torque situation, it was decided to change the gear box ratio from 150:1 to 200:1. This modification was made on VSR drive number one. Tests performed on the modified drive indicated that the pull increased to about 90 pounds, only $1\frac{1}{2}$ times the weight of the rod. Thus, the safety aspects of the rod will not be adversely affected.
2. Irregular brake frame movement caused difficulties in adjusting and maintaining proper clearance between the brake shoe and the brake disc. A slight drag on the brake further aggravated the rod withdrawal problem. Two small brackets were added to the frame of the drive unit. These brackets

keep the free arm of the brake frame centered. Additionally, a bumper block was attached to the frame. This bumper block counteracts the tendency of the brake frame to move when the brake is applied and should prevent the frame hinge pins from absorbing all of the brake load. These modifications have been made only to VSR drive number one.

3. The present rod in-limit switch is driven from the cable drum shaft and doesn't necessarily indicate that the rod is in (only that the drum is in the rod-in position).

The original design of the rod drives provides for a limit switch to indicate the rod-in position. Because of the high temperatures encountered at the bottom of the rod housing, this limit switch was installed in the drive assembly at the top of the housing and was driven by the cable drum shaft; therefore, it really indicates only the position of the drum. Several alternate methods were considered. One of these used a low pressure jet of nitrogen gas directed across the rod channel into a receiver tube to create a pressure in the receiver tube when the rod did not interrupt the jet. Several mockups of such a device were fabricated and tested to aid in the evaluation of the device. A 3/16 diameter tube appeared to be the best size for the jet when considering the amount and pressure of gas required and the pressure of the output receiver.

Of the several alternate methods considered, the nitrogen jet technique was selected because it has no moving parts in the rod housing, no electrical connections in the housing, is easy to fabricate and install, and is fail safe.

Specifications for all of the equipment required for these devices were prepared; a flow schematic of the piping system was prepared; and the jet units were designed and fabricated. One jet unit will be installed on the prototype rod housing in the 314 Building to check the techniques of installation.

VSR drive number one, incorporating the altered gear ratio and the brake frame brackets and bumper block, was reinstalled on the reactor. Its performance proved to be satisfactory both at room temperature and at high temperature. Modifications to the three remaining drives will be performed at the completion of the HTLTR hot tests.

Horizontal Control Rods. All horizontal control rod drives and elements were taken to the reactor building. Preliminary checkout of the rod drives under computer control was completed, and the rods were accepted by the Operations Unit.

A split ring was designed (10 rings are being fabricated) which will be installed on the actuating rod between the scram spring

bracket and the deceleration cylinder. Although this ring does not influence normal operation of the rod, it prevents the drive from over-traveling and damaging the rolling diaphragm or control element assembly in the event of failure of the in-limit switch.

Programmed Measurement and Control System (PMACS). Instrumentation and control lines between the reactor and control room were examined. Erroneous or troublesome ground connections were detected and corrected; the installation of electrical noise filters was recommended on the lines of several types of transducers.

Isolating transformers have been installed in the stepping motor outputs and the rod feedback pulses in the PMACS. They isolate the system electrically from ground. Noise filters have been installed on all low level analog inputs to PMACS. As a result of the isolation and filtering, the noise problem has been overcome, and successful operation of the analog measuring system under electrically noisy conditions has been accomplished.

A relay multiplexer test box was built and all multiplexer cards checked. About 5% of the original set of relays in the multiplexer were replaced because of their failure to operate (always open or always closed conditions were found on the relay contacts).

A brief feasibility study was performed of an inductive transducer system to be used as an "in-limit" position detector for the vertical safety rods. Initial information indicated that although the concept might be feasible, the material costs and delivery dates were unacceptable and a jet of nitrogen was selected to provide this limit indication.

Reactor Operations

The first part of the month was spent in preparations for the GAS CYCLE-HEAT design tests. Toward the end of the month the reactor heatup was begun. At the end of the month the average temperature of the reactor was 550 °C, with all equipment needed at this time working satisfactorily. The reactor heatup will continue until such time as a limit is reached on some system or some aspect of the reactor or equipment design. The work accomplished is given in more detail below.

The gas system was pressure-tested with helium and nitrogen, and was leak-tested with a helium leak detector. Several minor leaks were found and corrected. One significant leak at a weld on the reactor enclosure was found, and the project engineer was notified.

The gas chromatographs were calibrated and put into service. The moisture monitors were put into service. Some work, including calibration, remains to be done since they do not read correctly. Water vapor was monitored with a dry-ice dew point indicator during the reactor heatup.

All breakers in the electrical system were tested and adjusted for timing of their break point according to the recommendations given in an engineering review.

The devices which are relied upon to monitor power to the heater elements utilize the Hall effect. Their calibration was found to be in error, and considerable effort had to be devoted to their calibration. The control units which adjust the power to the heaters had to be recalibrated also. Since the problems were elusive and very subtle, two attempts were required, and a third effort will probably be needed to obtain the precision desired.

All heating rods were inspected prior to the reactor heatup, and all the connectors from the electrical buses to the heating rods were torqued to the same load.

The noise level on the thermocouple and other analog signal leads allowed accurate observations of the significant process variables to be monitored during the reactor heatup.

A surprisingly small amount of water was vented through the gas purge line. However, earlier indications of low residual moisture have been noted in previous reports. In large part, this is probably attributable to the change during design from diatomaceous earth insulation to a fibrous alumina-silica insulation. The moisture in the reactor was less than 1000 ppm most of the time, with high readings of about 3000 and low readings of about 200 ppm, depending upon the type of operation under test. The oxygen contamination was about 200 ppm after the initial evacuation and purge. The CO and H₂ contamination was barely readable during the month, and readings were in the range of 40 to 100 ppm. Continuous monitoring of hot spot temperatures and component expansions during reactor heatup is being done.

As a result of the operating experience gained during the month, several alterations were made to the computer programs and several bugs found.

The program, LIMIT, was altered to transfer all output alarms to Typewriter #2. The interference of the alarms from off-normal conditions on Typewriter #1 created unwanted interruptions on the control typewriter which created operating problems.

The heating system program uses the MAIN LOOP program to log the average temperatures in the four reactor stack segments. Considerable effort was spent finding out why the keyboard executive program, KEP, was locked out after the temperatures logs were transferred to magnetic tape. The problem was corrected.

A major bug in the gas system program was discovered and corrected. When the program state GAS-HEAT was requested and a low water cooling alarm occurred, the program should automatically change its state to COOL. However, the program jumped back to GAS-HEAT. A continuing alarm would cause cycling between the two states. This problem was eliminated by causing the program to lock itself into a requested state.

Detailed flow charts of the gas and heat system programs were completed, and portions of the document describing these programs were written. Several days were spent using the GAS-HEAT programs to help with debugging of hardware problems. The HEAT program proved particularly valuable for identifying heater problems.

At one point in the testing of the control rods power applied to the drive unit of one rod resulted in intermittent power to the other rods. This caused the other rods to "hunt" or oscillate in place. A subsequent, thorough investigation duplicated this condition and pointed to the likely cause. The potential for reoccurrence was eliminated. An intermittent short on an input to one of the rod drive units caused power to be put into the other eight units. They then oscillated, in place, until the short was removed, by blown fuses. No rod travel in either direction occurred.

Reactor Physics Program

Startup and Calibration Experiments. Following the testing of the oscillator and couplings in the hot tests in the HTLTR mockup, the final design of the graphite supports for the removable test cell, the graphite carrier for the normalizing poison sample, and the various kinds of oscillator-sample couplings was completed. Fabrication is proceeding.

Updated operating procedures were inserted at appropriate places in the document which describes the startup experiments. A review is now being made, and the document will be reproduced.

$U^{233}O_2$ -ThO₂ Experiments. Planning continued for the first U^{233} -ThO₂ experiments. Discussions were held with General Atomic to clarify the data most desired for the HTGR program and the experimental schedule which would be of most benefit to the program. Investigation of possible alternatives to the 1" diameter rods was pursued with the aim of proceeding towards lattices more like HTGR, i.e., fuel channels about 0.5" diameter. To construct lattices of this type in an economical manner, coated particles are practically a necessity and the possibility of obtaining such particles at this time is being reinvestigated.

The investigation of methods for calculating effective resonance integrals for Th²³²O₂ rods was continued. Use of the NR and NR1A approximation resulted in effective resonance integrals that are 10

to 20% lower than obtained experimentally for a number of ThC_2 rods by Palowitch and Hardy (Nucl. Sci. and Eng., July 1967). Application of Nordheim's integration scheme gave values that were in quantitative agreement with the experiments. The Nordheim's integration scheme has, therefore, been programmed into the current GAMTEC-II code and is presently undergoing debugging.

REACTOR FUELS AND MATERIALS

Fast Fuels Oxides and Nitrides (R. E. Nightingale)

Mixed Nitride Irradiations

Two experimental fuel capsules (GEH-14-745 and 746) containing mixed nitrides (UN-20 wt% PuN) continue to operate satisfactorily in the MTR. One thermocouple in GEH-14-746 has apparently failed, but readings from the other thermocouple plus those in GEH-14-745 indicate that the capsules have accumulated (as of June 19) a maximum exposure of 16,500 MWd/tonne. During their 42 equivalent full power days in reactor, heat generation rates have decreased ~13%; i.e., the heat generation rate for GEH-14-745 has decreased from ~32 kW/ft to ~28 kW/ft, while that for GEH-14-746 decreased from ~30 kW/ft to ~26 kW/ft. These capsules are scheduled to be irradiated to at least 20,000 MWd/tonne.

A third capsule, GEH-14-744, was charged into the MTR for a scheduled one cycle irradiation. However, because of reactor operating difficulties, the MTR ran at only 83% of full power during the entire cycle. This capsule therefore generated only 20 kW/ft while accumulating 3300 MWd/tonne, with a pin surface temperature of 440 °C. This capsule will remain in the reactor for an additional cycle during which it is anticipated that it will operate at 36 kW/ft with a pin surface temperatures of 650 °C.

The design for the remaining capsules in this experiment was modified slightly to provide more positive positioning of the thermocouples and to provide duplicate thermocouples in each position. Capsule components were fabricated and modified to incorporate these changes. However, no additional experimental capsules were fabricated because of difficulties in obtaining mixed nitride pellets of satisfactory quality.

Nondestructive testing techniques are being developed to assure high quality sodium bonds between the fuel pellets and cladding. As yet, no satisfactory technique is available. A computer study was initiated to determine the effects of imperfect sodium bonds on fuel capsule performance and to provide a basis for specifying sodium

bond quality. Initial results of this study indicate that a large void in the sodium bond can be tolerated without adversely affecting the safety or the value of the experiment. For example, a void from pellet to cladding, $\frac{1}{4}$ " long (one pellet) extending around 60° of the circumference of the pin would cause the maximum fuel temperature in these capsules to increase by no more than 60°C . With such a void, the maximum cladding temperature of the pin would be increased by less than 20°C .

Synthesis of Mixed Nitrides by a Carbothermic Reduction Method

Experiments on synthesis of uranium-plutonium nitrides and carbonitrides by carbothermic reduction of oxides in flowing nitrogen were started. The first objective of these experiments is to obtain data on the kinetics of the reaction:



A one-inch diameter molybdenum-wound tube furnace capable of 1800°C was used for initial experiments with UO_2 and carbon. X-ray diffraction analysis of a sample held in an alumina crucible at 1600°C for four hours showed only nitride and carbonitride patterns. The quantity of carbon actually required for complete UO_2 removal was about 10% greater than the theoretical quantity.

Measurements on reaction rates of UO_2 - 20 wt% PuO_2 - carbon mixtures were started using a graphite tube furnace with an alumina muffle and a recording carbon monoxide monitor. The powder was contained in a molybdenum and tungsten boat. One experiment at 1550°C was completed. A 10-gram quantity of feed material was 90% reacted in $3\frac{1}{2}$ hours. The reaction began at approximately 1250°C and gave a maximum carbon monoxide evolution of 14 cc/minute, corresponding to a conversion of 0.3 mole/minute mixed nitride. The reaction product was cooled in nitrogen. X-ray diffraction analysis showed that U_2N_3 was formed together with the mononitride (or carbonitride) phase. Lattice parameter of the mononitride phase was 4.892 \AA . Residual oxide was less than 2 wt%. Further chemical analysis of the product is in progress. This system can be easily adapted to a fluid-bed arrangement which is presently in the design and fabrication stage.

Work during the coming month will be directed toward establishing the temperature of maximum nitriding reaction rate.

Compatibility Experiments on Mixed Nitride Fuels

A sodium-bonded, 304 stainless steel-clad, UN - 20 wt% PuN test capsule heated to 650°C for 100 hours and two helium-bonded capsules, one heated at 650°C for 100 hours and the other at 1000°C for 100 hours, were opened and metallographically examined. No chemical

reactions were apparent, but there was some indication of mechanical spalling of the nitride pellet in the sodium-filled capsule. The outer 304 stainless steel protective tube showed partial oxidation at 1000 °C. It may be necessary to use a protective atmosphere for 1000-hour tests at 1000 °C.

One sodium-bonded capsule and one helium-bonded capsule are being heated at 650 °C for 1000 hours. Also, one helium-bonded capsule is being heated for 1000 hours at 1000 °C.

Two sodium-bonded, stainless steel-clad, UO_2 - 25 wt% PuO_2 test capsules (O/M ratios: 2.00 and 1.96) were cycled 285 times at 90-105 °C for a total time of 68 hours. Metallographic examination showed no chemical reactions or pellet spalling.

Additional capsule parts were fabricated and are now being assembled. High temperature testing (650 °C and 1000 °C) of specimens having O/M ratios of 1.92, 1.96, and 2.00 are planned.

UN - 20 wt% PuN Pellet Fabrication

Ball milling of UN - 20 wt% PuN powders prior to pressing and sintering is one of the more critical operations in mixed nitride pellet fabrication. An improved vibratory ball mill system of increased capacity has been evaluated, as described last month. Prior to placing the new system in the plutonium glove box, a pressure leak test was performed on the jars. Both jars leaked in the braze-bonded area of the steel jackets. Attempts to repair the jars by welding were unsuccessful. However, they were successfully sealed by machining grooves in the braze areas and filling with epoxy resin.

Stainless-steel ball-mill jars and balls were used for milling mixed nitride powders in an attempt to eliminate carbon pickup during milling. During a break-in run, using scrap sintered pellets, the mixed nitrides stuck to the stainless-steel jar and balled to such a great extent it was decided not to attempt further ball milling using that equipment.

Glass sample holders were designed and fabricated suitable for preventing oxidation of ball milled mixed nitride powders during transfer to the 200 Area analytical laboratory. The sample holders are designed to fit into a Perkin-Elmer Sorptometer for determination of specific surface area of powders. It is anticipated that surface area measurements will provide a means of correlating ball-mill parameters with sinterability of the ball-milled powders.

Considerable difficulty was encountered this month with the refractory metal resistance furnace used for sintering mixed nitride pellets. An air leak developed in the gas manifold used for back-filling the furnace. After that was detected and repaired, additional

poor sintering results were traced to the presence of diffusion-pump oil in the furnace chamber. Several batches of mixed nitride pellets were sintered during this period. Densities varied from 70 to 82% TD. Pellet densities of 85 and 90% TD are required for irradiation tests planned in the near future. Currently, considerable effort is being made to improve the purity of the atmosphere of the furnace during sintering.

Basic Swelling Studies (R. D. Leggett)

Irradiation Program

Construction and ex-reactor testing have been completed on capsule P-13 (700 °C, 5000 psi, 0.2-0.8 at.% BU). The status of capsules currently active is indicated below.

<u>Capsule No.</u>	<u>Control Temp. °C</u>	<u>Control Pressure (psi)</u>	<u>Goal Burnup (at.%) *</u>	<u>Status</u>
P-10	450	5000	0.2-0.8	Under irradiation
P-11	550	5000	0.2-0.8	Construction complete
P-12	625	5000	0.2-0.8	Construction complete
P-13	700	5000	0.2-0.8	Construction complete
P-14	700	500	0.2-0.8	Under irradiation
P-16	625	1000	0.35-0.7	Under irradiation

*Different burnups are achieved in a single capsule by including specimens of various enrichments.

The above irradiation capsules will provide data needed to evaluate the effects of temperature (alpha and beta phase), pressure, burnup, burnup rate, and minor alloying additives on the irradiation behavior of uranium.

Postirradiation Examination

Uranium and dilute uranium alloy specimens were recovered from capsule P-15 (700 °C, 1000 psi, 0.2-0.8 at.% BU). Each specimen was photographed in three different positions and the densities determined. Swelling values for specimens irradiated to ~0.2 at.% BU ranged from 5 to 15% per at.% burnup, specimens irradiated to ~0.4% BU swelled 10 to 15% per at.% burnup, and specimens irradiated to ~0.8 at.% BU swelled 15 to 20% per at.% burnup. Most of the swelling is thought to be due to cracking and tearing caused by operating close to and cycling through the alpha-beta transformation. The swelling observed in the 1000 psi test is not significantly different from the swelling observed previously in specimens irradiated at low pressure (~30 psi) and above 660 °C (beta phase). These previous specimens had "R" values of 7 to 16 at 0.1 at.% BU and 12 to 18 at 0.2 at.% BU.

Metallographic examination is being initiated to confirm the low swelling values and to determine the source of swelling.

High Pressure Postirradiation Annealing

Static, high temperature tests have been completed to ascertain the interaction between U, NaK, 304 SS, Ta, and Zr-2 at 900 °C. Specimens were inserted in three separate capsules, and special care was taken to isolate the samples from the 304 SS cans. This was accomplished by placing them in Ta baskets, suspended in the NaK. Three types of specimens were used--an uncovered cylinder of U, a similar cylinder but covered with 0.001" Ta sheet, and a quadrant of a Zr-2 clad U-U diffusion couple. The first two specimens were heated to 820 °C for 51 hours before a faulty temperature controller was discovered. The U-U diffusion couple was inserted into the furnace during the repairs, and all three specimens were raised to 900 °C for 96 hours. The weight changes observed in this test are:

Sample:	Pt 3 NA * <u>Uncovered</u>	Pt 3 NA * <u>Covered with 0.001" Ta Sheet</u>	Zr-2** Clad, U-U <u>Diffusion Couple Uncovered</u>
Net wt change: mg/cm ²	-49.6	-10.1	-30.7
Net Corrosion Rate:	-238	-48.3	-221

* At 900 °C for 96 hours and 820 °C for 51 hours.

**At 900 °C for 96 hours.

The general appearance of the samples and the 304 SS cans after the test was good except for some corrosion product (probably ZrO₂) on the Zircaloy. Three preliminary conclusions can be drawn from this experiment:

1. As long as U and 304 SS are physically separated, there appears to be no evidence of any eutectic formation. Hence, deleterious mass transport through the NaK does not occur in this system.
2. A relatively thin layer of Ta foil around the samples considerably reduces the corrosion rate.
3. Corrosion rates, although high, will not present any major problem for the swelling experiments anticipated.

It therefore appears that the dramatic interaction which was reported earlier between U and the other components in the capsule at 900 °C and 15,000 psi for 100 hours was entirely the result of unprotected U coming in contact with the 304 SS. Obviously, this is to be

avoided. To that end, three changes were made to the original capsule design. First, the U plug which was to be used as shielding inside the capsule was replaced with Ta. Secondly, the 304 SS sample holders which were previously lined with Ta foil were replaced with solid Ta sample holders. Thirdly, control samples of unirradiated U will accompany the irradiated samples to isolate any efforts which may be a result of the finite corrosion rate.

A new capsule has been constructed, tested, loaded with irradiated U, filled with NaK, and is currently being tested at 15,000 psi and 900 °C for an anticipated exposure of 100 hours.

Nondestructive Testing (J. C. Spanner)

Electromagnetic Testing Methods

The development of circuits for use in demonstrating the application of multiparameter test principles to a pulsed eddy current test is continuing. Construction of the multichannel eddy current tubing tester is nearing completion, and a laboratory evaluation of performance has begun.

Portions of the analyzer for the pulsed eddy current test include a second RC network which was wired to provide an additional signal to permit operation with a null, or near null, amplifier input signal, a four stage Laguerre polynomial generator, a capacitor discharge circuit for discharging integrator capacitors in the polynomial generator, a double pulse generator for use in providing timing pulses for signal sampling and capacitor discharge circuits. The double pulse generator circuit is the same as the one used to provide excitation for a simulator of ultrasonic nondestructive test signals. Next to be breadboarded and checked for operation with the polynomial generator are signal sampling and signal storage circuits.

The multichannel tubing tester is now an operating unit although further evaluation and final packaging remains to be done. Chart recordings of the two output channels have been made using an inside differential test coil assembly and 5/8" diameter x 0.050" wall stainless steel tubing. The tester was adjusted to discriminate against probe wobble signals on both output channels and to give a minimum signal on each channel resulting from a piece of aluminum placed against the outer wall of the tube.

Good separation was obtained between outer wall defect notches and inner wall defect notches 0.015" and 0.023" deep. As expected, less separation was obtained for deeper defects. For example, a region of suspected gross intergranular corrosion estimated to be about 0.030" diameter and 0.045" deep, starting on the inner wall of the tube gave a large signal on the inner wall channel with considerable residue showing on the opposite channel. Holes penetrating the tube wall gave similar signals.

Eddy Current Coil Design Studies

A study to investigate the theoretical and practical aspects of eddy current sensing coil design is continuing. Several experiments were conducted to investigate further the effect of tuning on probe sensitivity. The sensitivity of an encircling coil to a small drilled hole in a thin wall tube was measured as the tuning of the coil was varied through resonance. In one experiment it was found that the fractional impedance change due to the presence of the hole was approximately five times greater when the probe was tuned to resonance. In another experiment it was found that the output signal obtained from a commercial eddy current tester, while scanning the same hole, was increased approximately 2.4 times when the probe was tuned. Thus, it appears that significant sensitivity increases can be obtained by operating the probe coil at resonance.

Fundamental Ultrasonic Studies

The Kautz function analysis continued after errors which had been discovered in the EDPM program had been corrected. The program can now correctly calculate the first pair of Kautz functions to six significant figures, when this first pair is used as input data. This accurate reproduction stimulates confidence in applying the method of analysis to actual ultrasonic pulse data.

The above analysis and programming were also extended to include methods of converting spectral distributions to their corresponding ultrasonic pulses. Program subroutine "debugging" is still in progress.

It is anticipated that these latest developments will in the near future provide accurate representations of ultrasonic pulse behavior in attenuating media. A complete summary of the method and results will be forthcoming following the successful demonstration of the analysis technique.

Infrared and Thermal Research

Work directed towards improving the transducer used with the sinusoidal thermal wave tester continued. A viscous silicone rubber bonding agent was obtained for use in fabricating experimental solid backed thermal wave transducers. Tests showed that a good bond was obtained between the metal parts, but that bubbles were trapped in the rubber during solidification. The bubbles are thought to be caused from allowing the rubber to cure too rapidly. Tests are now being performed using smaller amounts of catalyst so that the mixture solidifies slower, allowing the entrapped bubbles to escape prior to applying the rubber to the transducer parts.

Assistance has been given to the AEC Chicago Patent Group in preparing a patent application on thermal transducers for non-destructively testing conductors, semi-conductors, and insulators (AEC Case S-31, 790-HWIR-1792). The application is expected to be submitted by July 1, 1967.

The Barnes R8T1 infrared radiometer was returned from the manufacturer after replacement of a faulty internal blackbody controller. Additional tests were carried out using the radiometer in the remote transient infrared nondestructive testing instrument. These tests indicated that the performance of the radiometer in this application had been improved. However, additional tests are needed to determine if the long term stability was improved sufficiently to permit extensive use on this project.

Considerable progress was made on the wide angle infrared imaging device (view field of $20^{\circ} \times 20^{\circ}$) being developed. This instrument, which will be useful in NDT research and other AEC applications, is now about 80% complete.

Composite Testing Methods

The compensation of effects of transducer characteristics on ultrasonic nondestructive test signals by use of signal processing circuits is being investigated. Circuits for processing a simulated signal were improved so that the duration of a relatively narrow band simulated signal was reduced to about one-fifth of its original duration. Application of the same circuit to signals from a fairly broadband 1 MHz ultrasonic transducer reduced the duration of the signal to about four-fifths of its original duration. The less effective compensation is due to the greater damping existing in the raw ultrasonic test signal as compared to that of the simulated signal. Compensating circuits having additional degrees of freedom will next be applied to the broadband signals to further improve compensation.

Detection of Radiation Induced Shift in Nil-Ductility-Transition Temperature

Mechanical and electronic apparatus has been assembled, and specimens are being irradiated for the purpose of developing an ultrasonic test method to detect radiation embrittlement in structural steels.

The electronic apparatus is being modified to enable tuning at preselected crystal controlled frequencies of 2, 5, and 10 Mhz. Wide band operation is still optional at other frequencies, but at reduced sensitivity. A linear RF gate circuit was also designed and fabricated.

Nine more steel specimens were prepared for irradiation during cycles 91 and 92 of the ETR at Arco, Idaho. Five of these will be irradiated under high temperature, medium high flux conditions, and four under low temperature medium flux conditions. These specimens complete the first family of irradiated samples which includes 28 specimens of three different materials representing seven exposure levels at low temperature, and 15 specimens of the same three different materials representing three exposure levels at high temperature. Ten temperature indicating specimens are also being irradiated along with the various specimen groups.

Nuclear Ceramics (R. E. Nightingale)

Materials and Information Exchange

Single crystal UO_2 specimens were prepared, characterized, and sent to the General Electric Company (NMPO) and to Oak Ridge National Laboratory (ORNL) for use in basic studies. The single crystal specimens included two large boules (80 g each), three spheres about 5 mm in diameter, and five discs 6 mm diameter by 1 mm thick. UO_2 single crystals enriched to 50% U-235, received recently from ORNL, were annealed in hydrogen for 12 hours at 1750 °C, cooled, and are now ready for the sphere grinding process. These spheres will be used in fission gas studies.

Nitrogen Analysis of Uranium-Plutonium Mononitrides

Promising results were obtained for nitrogen analysis in uranium nitrides by the Kjeldahl fusion method. Ten analyses by Kjeldahl fusion averaged 5.34% nitrogen, with an average deviation of ± 0.06 on a UN sample analyzed by vacuum fusion to contain 5.40%. Alumina, magnesia, and glazed porcelain crucibles were used in the analyses. The nitrogen analyses conducted in the alumina and magnesia crucibles averaged $5.37 \pm 0.04\%$ or about 96.7% of the theoretical nitrogen content of UN.

Uranium-Plutonium Nitride Synthesis Studies

Methods for preparing mixed uranium-plutonium nitrides by routes that do not involve reduction to the metals were investigated. Since the incentive for an alternate route is primarily economic, the material to be converted to nitride should be readily obtainable from nitrate solutions.

The reported preparation of uranium nitride by reaction of UF_4 with ammonia (at 500-800 °C) appears to be a promising method because UF_4 (and PuF_4) can be precipitated from nitrate solutions. A first attempt to prepare uranium nitride by reaction of ammonia with UF_4 did not succeed. X-ray diffraction analysis of the product

showed it to be a mixture of UO_2 and UF_3 . The formation of UO_2 indicates the presence of water and demonstrates that improvements in experimental technique are required.

Growth of PuO_2 Single Crystals

Installation of an electron beam zone-melting unit was completed, and a series of preliminary zone-melting experiments were made on sintered UO_2 rods contained in sealed tungsten capsules. The UO_2 was used as a standin for PuO_2 . Although melting was achieved on two occasions, controlled movement of a molten zone through a significant distance was prevented by the presence of defects in the tungsten tubing which allowed escape of vapor. This condition resulted in localized melting of the tungsten and gross escape of vapor leading to loss of vacuum and plasma discharge throughout the system. To attempt to remedy this difficulty, reactor-grade, vapor-deposited tungsten tubing will be utilized.

Nuclear Graphite (R. E. Nightingale)

EBR-II Irradiations

Property measurements were completed on the samples from BG-2 and BG-3, but the dosimetry is not yet complete. Temperatures indicated by melt-wire analysis agree with those estimated from heat transfer calculations. A fine grained, high CTE, isotropic POCO graphite that looked promising in BG-1 still has expanded only 0.1% at an estimated exposure of 10^{22} nvt at $\sim 650^\circ\text{C}$.

Pins are being fabricated to fill an entire 19-rod subassembly in EBR-II. Since some space is available in the core, approval for irradiation of entire subassembly in a Row 4 position has been requested. Exposures as high as 10^{22} nvt may be attained.

Irradiation of Nuclear Graphite

The last graphite irradiation capsule in this series, H-3-15, is still operating at design temperatures in the GETR. Seven of the original nine thermocouples are now operative. The capsule is scheduled to be discharged in July. The two reactor grade materials irradiated in this series, NC8 and CSF, will be included in the size-effect capsule currently being designed for the GETR.

Irradiation of "Proof Test" Graphites

Graphite irradiation capsule H-3-25 was discharged June 5, two weeks early, due to loss of thermocouples. The capsule was disassembled and the samples returned to PNL. Visual inspection revealed that no samples were broken or damaged but that turnaround had apparently occurred on several samples. Those samples with less

than 2 or 3% expansion will be inserted in the next capsule, H-3-26. Those with larger expansions will not be recharged but will be replaced with larger samples, 1.5" diameter by 2.25" long, to investigate the effect of sample size on dimensional stability.

All graphites irradiated in this program went through the sequence contraction-turnaround-growth. For irradiations in the temperature range 400-825 °C, all transverse samples turned around after maximum contractions of at most 1.5% and exceeded their initial lengths between 1 and 2×10^{22} nvt. At temperatures of 850 to 1275 °C, all transverse samples turned around after maximum contractions of less than 2.5% and exceeded their initial length before 8×10^{21} nvt. Parallel samples showed the same behavior and temperature dependence except that turnaround occurred at a higher neutron exposure.

Data points from all samples of a given orientation clustered together into two separable bands, one for irradiations at 400-800, and the other for irradiations at higher temperatures. Apparently there is a rapid increase in damage rate over the interval 800-900 °C.

Advanced HTGR Studies

Operating problems at the reactor have delayed charging of the first graphite irradiation capsule in the Dounreay Fast Reactor. The tentative schedule now calls for test startup approximately July 5, six weeks after the initial scheduled startup. Likewise, the charging schedule on capsules 2 and 3 will change by six weeks.

Effect of Oxidation on the Thermal Conductivity of Irradiated Graphite

Thermal conductivity measurements were completed on irradiated samples of CSF, TSX, and CSGBF graphite following increments of air oxidation. After 5%, 10%, 15%, and 20% oxidation, the conductivity at 200 °C was reduced 16%, 28%, 40%, and 50%, respectively. Similar reductions in the thermal conductivity at 400 °C were observed. The samples were oxidized in air at 550 °C. The exposure of the irradiated CSF, TSX, and CSGBF samples was 1.66×10^{21} , 1.6×10^{20} , and 2.16×10^{21} nvt, respectively.

Gas-Graphite Reactions

Microstructural changes taking place in TSX and AGOT-LS graphites as a result of reaction with oxygen under various experimental conditions are being studied. Initial experiments included rather mild oxidation attack at 600-800 °C and very aggressive attack in the glowing region of microwave-excited oxygen. Studies of the structural changes produced by oxygen in various conditions of reactivity which are intermediate between the two extreme conditions first investigated are now being pursued.

TSX and AGOT-LS microscope specimens were subjected to oxidation just inside the glowing region downstream from microwave-excited oxygen. The specimens were cut from graphite rod stock using a high speed alundum cutoff wheel. Oxidations were performed for periods of time up to 16 minutes at gas pressures of 100-200 microns and 500-1000 microns. The gas velocity in the reaction tube was approximately one meter per second.

Microscopic examination of the graphite specimens revealed that the rate of oxidation just outside the glow region is at least an order of magnitude lower than that in the glow region. Oxidation at the higher of the two pressure ranges is greater than that in the lower range in approximate proportion to the pressure ratio. The structural changes observed in the downstream region were considerably different from those produced in the glow region. No structures with straight edges or sharp angles, characteristic of the glow attack, were observed.

Fast Reactor Dosimetry and Damage Analyses

(R. E. Nightingale)

EBR-II Dosimetry Experiment

The flux monitor materials included in the second low power dosimetry experiment in the EBR-II were processed through the counting laboratory, and total flux was calculated for each monitor.

The reactor was run for one hour at a core power of 26 kilowatts. The monitor materials, which included 0.020" diameter wires of nickel, iron, aluminum, cobalt, and copper, were located in rows one, four, and six as in the first low power dosimetry experiment (reported in the May 1967 monthly report) but extended from the bottom of the core well into the upper blanket. Small pieces of scandium and depleted uranium were also irradiated at core midplane.

The EBR-II core consisted of 91 fuel and experimental subassemblies extending out through Row 6 during this experiment. The inner blanket, Rows 7 and 8, was stainless steel.

Calculated values of total flux in Rows 1, 4, and 6 at reactor midplane and axially along the Row 1 subassembly are listed in the table. Total flux was determined at each position from data from four types of monitors, $^{59}\text{Fe}(n,p)$, $^{58}\text{Ni}(n,p)$, $^{27}\text{Al}(n,\alpha)$, and $^{56}\text{Fe}(n,,)$.

Fluxes calculated from activation data of $^{59}\text{Co}(n,,)$, $^{63}\text{Cu}(n,,)$, and $^{45}\text{Sc}(n,,)$ disagreed substantially from the mean flux from the other monitors and was scattered. These $(n,,)$ monitors are potentially useful for fast reactor applications, but cross section uncertainties must be resolved before they can be relied upon for accurate dosimetry.

MEASURED EBR-II FLUXES

Radius, inches	Radial Flux Distribution at Reactor Midplane, $\text{nv/MW} \times 10^{-13}$				
	$^{54}\text{Fe}(n,p)$	$^{58}\text{Ni}(n,p)$	$^{27}\text{Al}(n,\alpha)$	$^{58}\text{Fe}(n,\gamma)$	Average
1 (Row 1)	3.94	4.58	4.04	3.85	4.10
7 (Row 4)	3.56	4.04	3.56	3.65	3.70
11 (Row 6)	2.10	2.31	1.92	2.89	2.30

Distance from Mid- plane, inches*	Axial Flux Distribution along Row 1, $\text{nv/MW} \times 10^{-13}$				
	$^{54}\text{Fe}(n,p)$	$^{58}\text{Ni}(n,p)$	$^{27}\text{Al}(n,\alpha)$	$^{58}\text{Fe}(n,\gamma)$	Average
- 6	2.94	3.46	2.92	3.04	3.09
- 4	3.42	4.15	3.46	3.37	3.60
- 2	3.75	4.48	3.85	3.65	3.93
- 1	3.92	4.56	4.10	3.69	4.07
0	3.94	4.53	4.04	3.85	4.10
1	3.88	4.50	4.00	3.83	4.05
2	3.75	4.37	3.65	3.75	3.88
4	3.31	3.90	3.15	3.56	3.48
6	2.60	3.08	2.50	3.17	2.84
9	1.85	2.02	---	2.79	2.22
13	1.25	1.46	---	2.12	1.61
17	0.692	0.827	---	1.31	0.943
21	0.385	0.500	---	0.962	0.616

* Core extends from -7" to +7".

Irradiation Damage to Reactor Metals (A. L. Bement)

Alloy Selection

A forged bar sample of a new Haynes alloy No. 561 has been obtained for evaluation. This high strength iron-base alloy is reported to have the following tensile properties at 1400 °F (760 °C), UTS 132 ksi, 0.2% σ_s 107 ksi, and elongation in two inches of 5%. An aging treatment lowers the strength slightly and doubles the ductility. The bar will be further reduced for fabrication into tensile and corrosion specimens for irradiation effects studies.

Several experimental vanadium-chromium binary alloys are being produced by Fabrication Metallurgy Section. Six experimental alloys in button form are being forged and rolled to a 0.010-0.020 thickness range for fabrication into mechanical test samples. A program is under design to investigate the effects of sodium on the creep properties of this alloy family.

The liquid metal capsule GEH 22-4 has been discharged from the ETR after approximately 50 full power days. The capsule functioned adequately, but a leak external to the capsule developed in the lead tube. Efforts are being made to determine the nature of the failure.

Capsule GEH 22-5 containing 38 nickel-base samples of alloy Hastelloy X-280, Incoloy 800, Inconel 625, In-120, and Inconel 718 is in final stages of fabrication. The specimens will be irradiated in a controlled temperature sodium environment with a sodium temperature range of 1100-1400 °F (593-760 °C). Goal exposure for this capsule will be 1×10^{21} fast fluence.

In-Reactor Measurements of Mechanical Properties

The purpose of the in-reactor measurements program is to determine the effects of irradiation on the mechanical properties of reactor structural materials. The effort is involved in measuring creep in AISI 304 stainless steel, nickel-base alloys, and refractory metal alloys during neutron irradiation.

An in-reactor creep test was conducted on hog swaged, sintered molybdenum at 580 °C and 21.1 kg-mm^{-2} (30,000 psi) stress. A creep rate of $2.4 \times 10^{-6} \text{ hr}^{-1}$ was observed for both the in-reactor test and the control test. The creep rate represents a portion of the transient stage creep since the creep rate of the control specimen is decreasing at the longer times. The in-reactor test ruptured after 431 hours, whereas the control specimen has not yet failed after 2000 hours. The elongation will be measured in the post-irradiation examination. Refractory metals such as molybdenum are very structure sensitive with respect to mechanical behavior. Work at GE-NMPO has demonstrated a large effect of irradiation on post-irradiation creep in vacuum melted molybdenum which had received a

high temperature anneal. Another test on molybdenum is planned in which a higher purity vacuum melted and annealed specimen will be used.

Irradiation Effects in Structural Materials

Stainless Steels. The purpose of this phase of the program is to determine the combined effects of irradiation and environment on the mechanical properties of stainless steels. Radiation-induced property changes will be determined from irradiations and tests conducted at various temperatures on several alloys. Particular emphasis will be placed on determining the existence of metallurgical instabilities and the mechanisms by which they are enhanced in a nuclear environment.

Transmission electron microscopy is being performed on annealed AISI 304 stainless steel tensile specimens that have been irradiated at 290 °C to a fast fluence of about 1.5×10^{20} n/cm² (E = 1 MeV), Quadrant 201. Observations of the buttonhead section of an as-irradiated specimen revealed defect clusters 50-100 Å in diameter. The defect density was about 1.5×10^{16} cm⁻³, and no depletion near grain boundaries or dislocations was seen. After testing at 400 °C, the clusters had increased in size and decreased in density. Dislocation loops were also observed at this temperature. Further studies will be performed on specimens deformed at higher temperatures.

A comparison of the irradiated and thermal control data for Quadrant 201 has been made. Radiation hardening of the yield strength was observed at 600 °C. Similar hardening of the ultimate tensile strength was significant only in the temperature range 200 to 400 °C. Unirradiated material exhibits a ductility peak at 450 °C, whereas irradiated material exhibits a minimum in ductility at this temperature.

Some 360 specimens of 304, 316, and 321 stainless steel in both annealed and 23% cold-worked conditions have been given various thermal treatments. These specimens are being prepared for use in a program to evaluate the effect of various pre-irradiation, thermal-mechanical treatments on the postirradiation properties of austenitic stainless steels.

Preparations are being made to irradiate part of the thermal-mechanical treated specimens in a graphite boat irradiation. The other irradiations will be carried out in the G-7 loop.

The mockup of the sheath capsule design (for irradiations at 400 to 600 °C in the G-7 loop) has been evaluated in Radiometallurgy. The thermal monitors employed in the capsules were removed and examined by x-ray diffraction. Diffraction peaks for the radioactive thermal monitors were sharp with no apparent blurring due to

the radioactivity. The monitors that melted during the irradiations indicate the temperatures in the capsules were in the range predicted by the STAC computer calculations.

Nickel-Base Alloys. The purpose of this program is to determine the effects of modified microstructures on the irradiation stability of nickel-base alloys. Microstructural modifications are made by pre-irradiation thermal or thermal-mechanical treatments and are evaluated by tensile tests, stress-rupture tests, and microstructural examinations.

A paper entitled "The Influence of Thermomechanical Treatments on the Strength, High Temperature Stability, and Microstructure of Hastelloy X-280," by I. S. Levy, has been accepted for presentation at, and publication by the International Conference on the Strength of Metals and Alloys to be held in Tokyo in September 1967.

Specimens of Inconel 600 in various experimental heat treatments were irradiated at 1250 °F (677 °C) to a fast fluence of 1×10^{20} n/cm² (>1 MeV). They, and their controls, were tensile tested at 1350 °F (732 °C). The data show that significant effects from both the thermal exposure and the neutron irradiation had occurred and that the magnitude of the effects is dependent upon the pre-irradiation treatments. Microstructural examinations performed on the control specimens confirm the above observations.

A comparison of the control data for the 1250 °F (677 °C) exposure with previously obtained 540 °F (282 °C) exposure data for Inconel 600 indicates that the standard treatment suffered an increase of 60% in yield strength and a 4-fold reduction in uniform elongation due to the exposure at 1250 °F (677 °C). An experimental treatment, on the other hand, showed only a 30% increase in strength and only a 28% reduction in uniform elongation.

Metallography explained these effects in Inconel 600. The 1250 °F (677 °C) exposure caused precipitation hardening in the standard treated material while the experimental treatment, which already had a significant precipitate structure due to the treatment, incurred less further precipitation and thus retained better ductility.

The effect of irradiation at 1250 °F (677 °C) on Inconel 600 was to further reduce ductility, though apparently not as a result of hardening since yield strengths were reduced 25% or more compared to controls. The standard treatment suffered a 5-fold reduction in uniform elongation (to 1.2%) and an 80-fold reduction in nonuniform elongation (to 0.6%), apparently the result of grain boundary embrittlement. The experimental treatment was more resistant to irradiation, ending up with 6.8% uniform elongation and 2.6% non-uniform elongation.

Stress-rupture tests at 1350 °F (732 °C) are continuing upon Inconel 600 and Inconel X-750 specimens irradiated at 540 °F (282 °C) to a fast fluence of 1×10^{20} n/cm² (>1 MeV) and their controls. The data to date indicate that irradiation reduces the rupture life of Inconel X-750 by 68% (427 versus 1386 hrs) and that of Inconel 600 by 74% (126 versus 490 hrs) in their standard-treated conditions. Some experimental treatments appear to increase the postirradiation rupture life for these alloys by about 25% (previous work on Hastelloy X-280 showed irradiation reduced rupture life of the standard treated material by 48% and that improvement of up to 107% could be achieved with some treatments).

Intercalibration Test of Counting Laboratories. Since the last reporting period, no other participating laboratories have reported their counting results of Pacific Northwest Laboratory.

Fast ($E > 1.0$ MeV) and thermal flux have been calculated for all monitor materials sent to participating laboratories. The spectral-averaged cross sections ($E > 1.0$ MeV) used to calculate flux from the Ni, Fe, and Ti monitors were 92.4 mb (branching ratio of 76%), 63.8 mb, and 10.2 mb, respectively. Helm's interpretation of the cross section measurements was used for the Fe and Ni monitors. The titanium cross section was determined by normalizing the flux from Ti to Ni. A value of 37.4 barns was used to calculate the thermal flux from the AlCo (0.1% Co) monitors. The average flux and the percent standard deviation from fifteen monitors of each type for both the 2-hour and the 8-day irradiation are listed in the following table.

Neutron Flux Calculated for MTR VH-2 Facility
at Reactor Mid-Plane

Reaction	Average Flux and % Standard Deviation for Each Monitor Set	
	2-hour Irradiation	8-day Irradiation
$^{58}\text{Ni}(n,p)^{58}\text{Co}$ ($E > 1.0$ MeV)	$2.36 \times 10^{13} \pm 2.8\%$	$2.44 \times 10^{13} \pm 0.8\%$
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$ "	$2.28 \times 10^{13} \pm 1.2\%$	$2.64 \times 10^{13} \pm 2.0\%$
$^{66}\text{Ti}(n,p)^{66}\text{Sc}$ "	$2.36 \times 10^{13} \pm 1.2\%$	$2.20 \times 10^{13} \pm 2.0\%$
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ (Thermal)	$1.79 \times 10^{14} \pm 2.1\%$	$1.54 \times 10^{14} \pm 2.9\%$

Discrepancies are noted in the above table between the low and high exposure flux levels. Mr. C. H. Hogg of Idaho Nuclear Inc., who arranged for the irradiation of these materials, indicated that the differences between the low and high exposure flux levels might be explained by the following reasons:

1. Variation of power level;
2. Instability of water pressure in rabbit facility, thus causing capsule to drop from mid-plane position; and
3. Disintegration of aluminum foil spacer in 8-day irradiation causing a shift in positioning of the foils in the capsule.

However, the absolute value of the flux is not important in this study, but only the differences between individual laboratories.

In-Reactor Corrosion and Hydriding of Titanium, Niobium, and Zircaloy-2. Specimens of Grade II titanium and high purity niobium were exposed in the G-7 loop of the Engineering Test Reactor in pH-10 NH_4OH , <0.05 ppm O_2 at 270-280 °C. The length of the exposure was four ETR cycles, 84 days at temperature. The estimated flux was 1×10^{14} n/cm² sec, 1 MeV, and the estimated fast fluence is 7×10^{20} n/cm².

The in-flux and out-of-reactor weight gains and hydrogen pickups are shown in the table for titanium and niobium; Zircaloy-2 is included for comparison. Titanium and Zircaloy-2 were exposed as etched; niobium surfaces were electropolished before exposure.

In-Flux and Out-of-Reactor Weight Gains
and Hydrogen Absorption

Specimen	84-Day Weight Gains		Hydrogen Pickup			
	In-Flux	Out-of-Reactor	In-Flux*		Out-of-Reactor**	
	mg/dm ²	mg/dm ² ***	ppm	mg/dm ²	ppm	mg/dm ²
Titanium	44.4	0.7	151	3.4	0	0
(Grade II)	43.6					
Niobium	84.1 88.9	145	13	0.7	20	1.1
Zircaloy-2	92.5 98.5	12.2	224	4.2	42	0.8

*After 84 days in-flux.

**After 200 days in out-of-reactor autoclave.

***Autoclave.

Data were reported in the January 1967 monthly report for samples of the above materials exposed two ETR cycles (37 days) in the same system. Titanium and Zircaloy-2 weight gains and hydrogen pickups are much higher after the 4-cycle exposure. Weight gains for niobium were nonreproducible after the 2-cycle exposure. After four cycles, weight gains were reproducible and of the same magnitude

as the maximum weight gain after two cycles. The niobium in-flux weight gains are lower than the out-of-reactor weight gain, suggesting that oxide loss may have been accelerated by the flux. In-flux hydrogen pickup for niobium was low relative to values for titanium and Zircaloy-2.

The weight gain for etched Zircaloy-2 is higher than previous G-7 loop exposures in pH-10 NH_4OH would predict, for reasons which are not apparent.

Fast Reactor Studies

Structural Materials and Fuel Cladding Studies. The objective of this program is to determine the combined effect of environment and fast reactor irradiation on the mechanical properties of candidate fast reactor cladding and structural alloys. The program currently in progress is specifically directed at providing a basis for selection of optimum alloys for FFTF applications and at providing a description of material behavior in fast reactor service.

Tensile specimens of AISI 304 and 348 stainless steel in both the annealed and 25% cold-worked conditions irradiated in the EBR-II to 1.7×10^{22} n/cm² total neutron fluence are currently being tensile tested. These specimens were fabricated from the same material as was used on the "Irradiation Effects to Stainless Steel" program which emphasizes irradiations in the ETR. Thus, the results from the EBR-II irradiations on this material will provide a basis for comparison of the damage states produced by the two reactors. The tensile testing is nearly 75% complete, and results are being tabulated.

Apparatus for postirradiation creep tests on annealed AISI 304 stainless steel irradiated in the EBR-II to a peak dose of 1.7×10^{22} n/cm² is being calibrated at BMI-Columbus. Control creep testing is in progress at PNL.

Samples of 304 stainless steel irradiated in the EBR-II to 1.7×10^{22} n/cm² (fast) have been further analyzed by transmission electron microscopy. The Frank sessile loops transform into perfect loops in the temperature range 1100-1300 °C. The Burgers vector of the perfect loop was determined as $\frac{a}{2} [110]$. At 1200 °C, there are still some faulted loops remaining;² at 1300 °C, all loops have transformed and the loops have further developed into a nodal structure. The Burgers vectors of the nodes were analyzed and found to be consistent with the reaction:

$$\frac{a}{2} [110] + \frac{a}{2} [\bar{1}01] = \frac{a}{2} [011].$$

At annealing temperatures greater than 1300 °F (704 °C), the dislocations anneal out by typical recovery processes. No evidence of gross grain boundary migration, i.e., recrystallization, was found.

Thin sections, cut parallel to the sample axis and near the fracture end of a tensile sample, are being studied. The microstructure is characterized by a high density of dislocations similar to that expected in a highly deformed unirradiated sample. The one significant observation so far is the randomness of the voids. There is no evidence of dislocations sweeping up bubbles during deformation.

Fast Neutron Mechanisms. Fundamental studies of hydrogen behavior in fast reactor alloys are continuing using both dynamic and steady-state hydrogen permeation techniques. During the month a new permeation facility incorporating a high sensitivity, mass spectrometer type hydrogen leak detector was assembled, calibrated, and placed in operation. The usable hydrogen sensitivity for permeation experiments is greater than 2×10^{-7} atm cc/sec for normal hydrogen backgrounds, while the time constant for pressure changes in the permeation system is less than 0.25 sec.

A series of hydrogen solubility and gas evolution studies using the new detector system has been initiated to determine the effects of irradiation and prior thermomechanical treatment on (1) the occlusive capacity, and (2) the isothermal and isochronal hydrogen evolution characteristics of fast reactor alloys in the 300-700 °C temperature range. Planar specimens are charged with hydrogen, quenched, and the occluded hydrogen vacuum extracted under isothermal or isochronal conditions. Integration of the resulting evolution rate versus time/temperature curve gives the quantity of gas occluded for the particular charging and specimen conditions involved. The use of this procedure eliminates a number of the problems associated with the conventional Sieverts technique and permits hydrogen evolution rates (for the approximately 0.5 g samples used) as low as 0.0002 ppm/sec to be detected.

A preliminary comparison of the isochronal hydrogen evolution characteristics of representative hydrogen-charged AISI 304 and 316 stainless steel samples has been completed. For the test conditions employed ($dT/dt = 24$ °C min), the maximum hydrogen evolution rate for AISI 304 stainless steel was approximately 60% greater than that for AISI 316 stainless steel, and the temperature at which the maximum occurred was significantly lower (485 °C versus 520 °C). The effect of hydrogen charging conditions was also investigated, and a significant difference in isochronal evolution behavior was noted for specimens thermally charged with hydrogen and those charged as the cathode in a high intensity, hydrogen-helium glow discharge.

Thin Section Materials. The program to select a material for an in-core pressure device for the FFTF is continuing. (Previous reports appeared under Alloy Selection.) The 0.010-inch-thick sheet specimens have been machined. They are ready for annealing. The 0.015-inch-thick sheets of 304, 316, and 348 are being sheared into specimen blanks in preparation for machining.

The investigation of the corrosion film on the 304 and 316 specimen subjected to 1060 OF (571 OC) sodium for 609 hours in the isothermal loop is complete. The metallographic examination showed the film to be extremely thin such that it could only be resolved at 1000X. The results from x-ray diffraction indicate there is a body-centered-cubic structure present that could be either alpha iron or chromium. The results from the spectrographic analysis indicated the manganese was higher in the surface scraping than in the bulk alloy. The results from the microprobe indicate the film was made up of the elements Na, O, and Cr. The true nature of the corrosion film is still not known. From the information available at least two mechanisms could account for the weight gain experienced by the specimens. The first possibility is that chromium and manganese have formed on the surface of the stainless steel specimens in a deposition process with the sodium and oxygen present as a contaminant left over from the cleaning process. The second possibility is the corrosion film is formed as a mixed Na-Cr-Mn oxide that is adherent to the surface of the stainless specimen with the Cr and Mn coming from the stainless steel.

There is a question as to the operating conditions in the isothermal loop being compatible with the needs of the program. The needs of the program and the operating conditions are being evaluated to determine if further use should be made of the isothermal loop in this area of investigation.

Test Facilities. A liquid metal metering station for handling high purity sodium has been assembled and tested. This station makes possible the transfer of measured volumes of sodium from shipping and storage vessels to receiving vessels without danger of contamination. A formal operating procedure has been prepared and submitted for safety approval. Calibration tests have yielded an average error of 1 ml with a maximum error of 3 ml in a volume range of 220 ml (minimum) to 500 ml. With present equipment, volumes less than 220 ml must be delivered with the aid of an inert atmosphere glovebox. A sample of sodium may be taken before or after delivery of the desired volume for an analytical check on the purity of the supplied sodium.

Bids for the liquid metal storage and loading facility, which will include an inert atmosphere glovebox, are expected late this month.

Design for renovation of 321-A Building is about 70% complete.

The support system for the sodium loop, wherein a flowing sodium test environment will be maintained, was reported ready to ship on June 2. With normal delivery time, this equipment should be delivered early in July.

ATR Gas Loop Supporting Studies

Model Gas Loop Studies. This loop is a one-tenth size model of the gas loop that is being constructed for the Advanced Test Reactor at Idaho Falls. It is a recirculating helium loop, rated for high temperature, 2100 °F (1149 °C), which is being operated to determine the effect of helium on high temperature materials and specimen holders, and to assess the effects of prolonged use on the operating characteristics of the new heater.

The loop has operated a total of 3935 hours since installation of the new heater, with 75 thermal cycles from room temperature to various heater outlet temperatures. During the month, repairs were made to several system components as follows:

1. The French motor-generator variable speed drive unit which has given considerable trouble in the past was removed and disassembled. Grease and water were found within the internal parts; a grease seal is being installed to eliminate this problem. The six-month period of operation of this unit without bearing failure indicates that previous lubrication problems are solved.
2. The three-stage compressor of the helium purification system failed and has been repaired and put back into service. A chromed cylinder was added to the first stage.

An addition to the horizontal test section has been designed and is being fabricated. This will enable loop startup with the test specimen in a cool zone about 100 °F (38 °C) until high temperature, 2100 °F (1149 °C), has been obtained with high purity helium gas. When desired operating conditions are reached, the test specimen can be remotely moved into the high temperature zone. The design will allow retraction of the specimen into the cool zone when desired. The operation of this system will be evaluated when completed, and the loop will be placed in operation shortly after July 1, 1967.

Damage Mechanisms in Iron

The objective of this program is to determine the interaction of defects produced in iron by neutron irradiation with moving dislocations which gives rise to the changes observed in mechanical properties.

During this period the major effort was directed toward completing a paper entitled "The Analysis of Thermally Activated Flow in Alpha Iron Using Temperature Change Techniques." This paper will be submitted to Acta Metallurgica for publication.

Equipment development for resistivity measurements and ultrasonic attenuation measurements has continued. Data from these tests should be available in the next quarter.

High Pressure Studies

Initial studies with the diamond high pressure x-ray cell have revealed that the actual pressure experienced by a sample enclosed with a metallic gasket is a sensitive function of the sample-gasket compressive strength ratios. Experiments with Zr, InTe, Ce, CdS, RbCl, ZrO₂, Gd₂O₃, and Nd₂O₃ show that the sample pressure is ~0.8 the theoretical pressure (force/area) for metals and ~0.4 for ionic and covalent compounds. This agrees with a previous theoretical analysis (BNWL-438). The implication of this study is that workers in the field may overestimate the sample pressure by a factor of 2 or more with some compounds and by 25% on metals if they assume that the pressure is simply force/area.

In the course of these studies the density changes associated with the pressure-induced phase transitions were measured for Zr, InTe, Ce, CdS, RbCl. In addition, a transformation in Gd₂O₃ from a cubic to monoclinic structure was noted at 40 kbars and 22 °C. This transition has not been reported in the literature. Hexagonal Nd₂O₃ showed no phase transition up to 50 kbars as was the case in monoclinic ZrO₂ up to 90 kbars at 22 °C. It had been previously predicted by other scientists that this latter transition would take place at 58 kbars and 22 °C.

With a better knowledge of the pressure actually experienced by a gasketed sample, the first set of experiments to measure the combined effects of irradiation and pressure on phase transition has been initiated. Zr metal was doped with 1000 ppm of natural uranium and oxidized to ZrO₂. The reaction of the doped ZrO₂ to pressure is being ascertained, and then this sample will be sent to the reactor, still under pressure, along with a sample at 1 atm. The difference in the amount of conversion to the tetragonal phase will be noted.

The test of the in-reactor hydrostatic pressure cell which had run at 17 kbars and 22 °C for 90 days was stopped. Upon unloading, the inner cylinder cracked, allowing some sample to be extruded. The reason for this cracking may be associated with the rate of stress release or due to binding ring compressive stresses which act as the internal pressure is released. Both of these possibilities are now being studied.

The ex-reactor belt and piston-cylinder units have been used to prepare large quantities of the pressure polymorphs of Zr, InTe, PbF₂, PbO₂, SiO₂, and P. X-ray analysis of all of these materials before and after treatments at pressures up to 60 kbars and 1000 °C show that the high pressure forms have been produced and quenched to room conditions. The changes in some of these materials are quite dramatic, involving density increases of 2% (Zr) to 50% (P) over the theoretical density of the starting materials. Color changes have also been noticed; black to blue for InTe, white to

to black for P, and red to black for PbO₂. Attempts to quench a metallic form of CdS back to 1 atmosphere were not successful, and the conversion of silicic acid to coesite was not as large as anticipated.

The high pressure polymorphs produced will be irradiated to a fast fluence of $\sim 5 \times 10^{18}$ n/cm² and subsequent hot x-ray analysis performed to see if the increased density can be retained.

ATR Gas Loop Operation and Maintenance (G. A. Last)

ATR Gas Loop Support

Vendor Data Review. Vendor data for the ATR gas loop are being reviewed. The majority of the data currently being submitted are corrected data based on past comments by the various reviewing agencies.

Analytical Instrumentation. A meeting was held with the contractor for the gas analytical instrumentation to review the unresolved engineering details. Preliminary inspection procedures were reviewed, and correlation with the detailed inspection procedures will be prepared by PNL.

The chromatograph and total impurity analyzer currently being tested by PNL are in the process of preparation for shipment to Idaho Falls.

Hastelloy X Pressure Piping. A meeting was held at PNL with Ebasco and AEC-ID personnel to review the pressure and thermal stresses on the high temperature Hastelloy X piping. The review concluded that the piping stresses were within code requirements, but due to the limited experience with Hastelloy X, a surveillance program will be initiated. In addition to the surveillance program, PNL will do further testing on samples of the actual piping to assure that the materials have the mechanical properties required for ATR gas loop service.

Regenerative Heat Exchangers. The original design submitted on the regenerative heat exchangers used a combination of stainless steel and Hastelloy X. During the review of vendor data it became apparent that an all-Hastelloy X unit would offer more reliability in the ATR gas loop. PNL has a large inventory of Hastelloy X billets, and the required nondestructive testing, stress rupture tests, etc., have been completed to determine that the material will meet all of the requirements for its intended service. This material will be shipped to the manufacturer of the heat exchangers to expedite delivery of the final product.

Metallic Fuels Development (G. A. Last)

Irradiation of Thorium-Uranium-Zirconium Fuel Elements

Three thorium - 2.5 wt% uranium (93.2% U-235) - 1.0 wt% zirconium tubular fuel elements clad in Zircaloy-2 are under irradiation in the P-7 loop in the ETR. Three fuel elements which were being irradiated are being stored in the viewing basin at the ETR site.

The current status of the six test elements in this test is summarized in the following table. The behavior of the elements continues to be excellent with only 3.4% swelling being observed on the element with the maximum exposure of 2.1 at.% BU.

These elements are presently being transferred to a hot cell for determining their volumes and physical dimensions during ETR cycle 90. Three of the elements will continue their irradiation in the P-7 loop during subsequent ETR cycles.

GEH-10	Percent Fuel Swelling	Burnup		Max. Core Temp. °C	Spec. Power w/gm	Surface Heat Flux BTU/Hr/Ft ²
		Fissions/cm ² (MWd/tonne)	At. %			
65*	3.4	6.3 x 10 ²⁰ (18,600)	2.1	460	39.0	5.7 x 10 ⁵
64*	1.7	4.4 x 10 ²⁰ (13,000)	1.5	420	30.7	4.5 x 10 ⁵
71	1.3	3.2 x 10 ²⁰ (9,400)	1.1	480	44.2	6.4 x 10 ⁵
72**	0.1	1.5 x 10 ²⁰ (4,300)	0.49	490	43	6.2 x 10 ⁵
70**	-0.5	1.3 x 10 ²⁰ (3,900)	0.45	470	39	5.6 x 10 ⁵
84**	0.1	0.9 x 10 ²⁰	0.29	440	33	4.8 x 10 ⁵

*Temperatures, corrected for oxide buildup, and heat generation conditions during ETR cycle 89.

**Temperatures, not corrected for oxide buildup, and heat generation conditions during ETR cycle 84.

High Exposure Uranium Irradiation Test

Hollow core uranium fuel elements being tested in the M-3 hot water loop of the ETR have successfully completed their fifth cycle of irradiation. At the end of the fifth cycle, ETR cycle 89, the accumulated exposures ranged from 5143 MWd/tonne to 394 MTd/tonne. The maximum fuel temperatures range from 565 °C to 350 °C. These fuel elements, 0.45-inch diameter by 6.25 inches long and clad with Zr-2, are part of an irradiation test that has been designed to operate uranium fuel rods at elevated alpha phase temperatures to burnups greater than 10,000 MWd/tonne (6×10^{20} fissions/cm³). The variables being studied in this test include fuel composition, external restraint, and internal void volume. The combined effects of the plastic character of uranium during irradiation and the restraints from the cladding and system pressure are expected to cause the uranium swelling to be accommodated by a central hole. Two uranium compositions are being used: Alloy 1, containing U + 350 ppm Fe - 800 ppm Al, and Alloy 2, containing U + 150 ppm Fe - 100 ppm Si. A fuel enrichment of 4.5% U-235 is being used to achieve the desired burnup rate (10,000 MWd/tonne in one calendar year) and temperatures. Fuel rods of both compositions were fabricated by coextrusion to 0.450 inch diameter with 0.025 inch and 0.050 inch thick Zr-2 cladding.

At the end of cycle 89 weight measurements were made in the reactor basin on the 24 fuel elements in the test assembly. Volume changes calculated from these weight measurements showed that all of the fuel elements continue to show small volume decreases. The data continue to show a reversal of the volume decrease that is dependent on the initial internal void volume and on the cladding thickness. Extrapolation of the volume change data for the 0.050-inch Zr-2 clad elements with 10 and 20% nominal void volumes indicates that an exposure of 7500 MWd/tonne will be achieved before the fuel volumes return to their preirradiation values. Warp measurements were made on each of the 34 elements thus far irradiated in the test. Five of the elements have a double throw warp between 0.05 and 0.1 inch. The remaining all have double throw warp less than 0.05 inch.

Negatives from neutron radiography have been received on eight fuel elements shipped to the Battelle Research Reactor at the end of cycle 88. Seven of these elements had been neutron radiographed at the end of cycle 86. Between cycles 86 and 88 the volume decrease had stopped and reversed on two of these elements. It was anticipated that the stop or reversal of the volume decrease might be associated with the filling or near filling of the central void. However, the neutron radiographs revealed only small decreases in diameter of the central voids. The neutron radiographs also revealed transverse cracks in the fuel of five elements. Examination of the neutron radiography negatives obtained after cycle 86 also

showed cracks in the fuel of two elements. These cracks can be seen in the negatives taken after cycle 88, indicating that the cracks did not heat on continued irradiation.

Fabrication of 12 additional fuel elements for this test has been completed through the brazing step. These elements will be used for irradiation testing to goal exposures beyond 10,000 MWd/tonne.

Hollow Zr Clad Uranium Rod Fabrication

A highly successful experiment was performed to determine the feasibility of coextruding 0.600 inch diameter, 0.025 inch Zr clad, hollow uranium rod in which the void volume is as low as 5%. These dimensions require a hole in the uranium approximately 0.12 inch in diameter. Two billets were fabricated to test two approaches for obtaining these dimensions: (1) extrude directly to 0.12 inch diameter hole using a solid mandrel and (2) extrude, over a solid mandrel, to a larger hole and reduce the hole to 0.12 inch diameter by swage sinking. The billets were 2.25 inches OD by 4 inches long with 0.125 inch and 0.250 inch diameter holes, respectively. Both consisted of 0.035 inch wall copper on the outside, a 0.095 inch wall Zr-2 sleeve, the uranium, 0.010 inch wall copper on the inside, and 0.125 inch thick electron beam welded copper end caps. The floating mandrels were made of T-1 tool steel with a 0.010 inch diametral taper in the working length. The small hole billet was extruded through a 0.625 inch diameter die and the other through a 0.750 inch diameter die.

The billets were lubricated with Aqua Dag and preheated to 620 °C and the tools, with exception of mandrels, were lubricated with Oil Dag and preheated to 500 °C. The mandrels were neither lubricated nor preheated. Both extrusions were successful with the holes being of uniform diameter, centered, and of excellent finish the full length.

ENGINEERING DEVELOPMENT

Neutron Flux Monitors (W. G. Spear)

Regenerative Detectors (Thermal Neutron Flux)

Accelerated preparations furthered progress on the comprehensive reactor in-core test program regarding evaluation of the characteristics of the new regenerative thermal neutron flux detectors. A total of 10 detectors, including regenerative, fission, gamma, and self-powered beta-current devices, are being assembled into a single test capsule for irradiation at the ETR. Completion of the electronic

instrumentation system will permit accurate, comparative measurements of the operational characteristics of the detectors during the planned, extended evaluation test. Assembly of the test capsule progressed following acquisition of all major components and the various detectors.

In the regenerative detectors the usual rapid burnout problem at high neutron flux levels for the normal U-235 detector is overcome through use of the developed U-234 - U-235 regenerating technique. In this method the U-234 material contained within the detector transmutes to U-235 to provide replenishment of the fissioned atoms to assure long-lived, proper operation during reactor in-core use.

Microwave Detectors (Thermal Neutron Flux)

Ionization of a gas contained within a microwave cavity will cause the resonant frequency of the cavity to shift by an amount proportional to the average free electron density. If this ionization is caused by the charged particles from the (n,p) reaction of thermal neutrons with He-3 or by the fission fragments from U-235 or by a mixture of isotopes such as the regenerating mixture of U-234 and U-235, then the frequency shift will be proportional to the neutron flux density. Thus, this technique can be very useful for measurement of reactor in-core neutron flux.

During previous experiments a significant reduction of sensor sensitivity was noted. This reduction of sensitivity has been attributed to the use of a quartz capsule used to contain the gas inside the microwave cavity. Recent calculations indicate that cavity electromagnetic fields are perturbed by inclusion of a quartz capsule. For a cavity, assumed to operate in the TE₀₁₁ mode (i.e., the electric fields transverse to the longitudinal axis of the cylindrical cavity), the electric field tends to concentrate in the dielectric. Assuming a given incident thermal neutron flux, hence, a given plasma free electron density, the calculated frequency shift of a dielectric-containing cavity is less than one-tenth that of a dielectric free cavity. Thus, the development of a dielectric free cavity is expected to increase the sensitivity significantly. Several cavities with metal-to-ceramic seals have been designed to provide the desired gas tight cavity without the use of a quartz capsule. Techniques to obtain suitable metal-ceramic seals in waveguide sections have been successful. One cavity has been assembled using a relatively low melting point solder (185 °C), and two others are being assembled elsewhere following rigid specifications. These cavities will use He-3 and will have gas pressure variation capability.

Regenerating Detectors (Fast Neutron Flux)

Rugged, long-lived, reactor in-core fast neutron detectors are needed for use in advanced reactors where severe temperature problems, high fast flux levels, and extreme gamma levels prevail. It appears that suitable regenerative techniques may provide the appropriate performance characteristics needed to achieve accurate fast neutron flux measurements under the anticipated reactor operating conditions.

The capture and fission characteristics of possible candidate isotopes are under investigation to establish suitable combinations applicable for development of a fast neutron regenerative detector. Computer analysis methods are being used to explore the pertinent cross sections of particular fertile and fissile isotopes.

Beta Current Generator Detectors (Fast Neutron Flux)

The Be-9 isotope shows promise as a possible detector material for measurement of fast neutron flux under the difficult environmental conditions anticipated in future advanced reactors. This isotope undergoes an (n, α) reaction, with a 0.1 barn cross section at 3 MeV, to provide a beta particle emission (\sim 3.5 MeV) with a relatively short half life to 0.8 second. The beta emission generates a signal current proportional to the neutron flux level to yield an estimated signal current of approximately 1×10^{-6} A from about one cm³ of Be exposed in a fast neutron flux of 10^{15} nv. In an experimental chamber beryllium in a metal oxide state can be encapsulated in a suitable sheath opaque to the alpha particles but transparent to the generated betas, which would escape for signal generation. To avoid interference signals due to the anticipated high gamma levels of 10^{10} R/hr, the balanced twin-lead, twin-chamber concept, as developed during the thermal flux detector investigations, should prove useful.

Evolved Gas Detection Concept (Fast Neutron Flux)

In this unique concept for advanced reactor fast neutron flux detection an evolved gas, such as helium, would be collected through a simple evacuation tube connected to a chamber assembly inserted into the reactor core. Minimized gamma interference could be achieved by location of the electronic instrumentation in the less destructive environment external to the core. In the detector chamber (n, α) or (n,p) reactions with B-10 or other materials would release gas in quantities proportional to the neutron flux density. Calculations indicate that about 10^{11} atoms/sec of helium gas would evolve from one gram of B-10 in an incident fast neutron flux of 10^{13} nv. At these levels the evolved gas could be measured by mass spectrographic or optical spectrometric methods. It is expected that neutron flux levels down to 10^{10} nv could be detected using the evolved gas concept.

Microwave Detectors (Fast Neutron Flux)

By extending the microwave techniques being developed for use in reactor thermal neutron detection, it is anticipated that reactor fast flux may be measured. A cavity containing a material with an appreciable cross section to fast flux, such as U-238, could be used. It may also be possible to use the proton recoil in hydrogen as a source of ionizing particles. The gamma-caused ionization in the cavity could prove troublesome, thus development of compensation techniques will be necessary. One such technique is to measure the frequency difference between a microwave generator, which is locked electronically to a reference cavity, and the sensing cavity for the fast neutrons.

Other neutron initiated reactions causing frequency shift or energy absorption are also being considered. For example, materials such as Be-9 which evolve helium gas in their reactions could be used to inject their evolved gas into a cavity thereby changing the frequency. The sensitivity of such a sensor would be related to the gas evolution rate. For example, for a gas evolution rate of 10^{-2} A sec^{-1} nv^{-1} , fast neutron flux measurements down to about 10^{11} nv could be achieved.

Microwave and Infrared Detection of Coolant Impurities and Measurement of In-Reactor Temperature (W. G. Spear)

Microwave Detection of Impurities in Coolant Gases

The propagation properties of electromagnetic waves in structures such as waveguides and cavities are dependent upon, among other things, the dielectric constant of the material contained within the structure. The dielectric constant of a gas mixture is a function of the number of molecules of each gas present and the molecular polarizability. Thus, as water vapor is introduced into an otherwise pure gas such as helium, the dielectric constant changes. This change in dielectric constant has been used to cause variation of the frequency of a microwave cavity. In this manner a detection sensitivity of 125 ppm of water vapor in helium at 300 °C and of 950 ppm at 436 °C has been achieved. The time response of the detector is small, being limited by the time constant of the electronic measuring apparatus. Time constants of one second were used in the experiments. Greater sensitivity can be achieved by using a very stable microwave oscillator and heterodyne techniques. It is also apparent that for appropriate sensitivity at very high temperatures, it may be necessary to cool the gas sample to some extent.

A second promising method for such measurement involves the use of microwave phase shift in a section of waveguide. The section could be included as an integral part of the reactor process tubing. Preparations are under way to perform experiments to establish the

sensitivity of this method and to explore the feasibility of using large tubing as "waveguides" for the microwave frequencies used.

High Temperature Measurements

For many nuclear reactor designs, temperature of constituent materials limits the maximum operating power level. Consequently, accurate and precise measurement of reactor in-core temperatures permits operation of the reactor optimally. Several microwave temperature sensors are being investigated for their application in a high temperature, radiation environment.

Successful operation of one type of sensor, made of copper and Carpenter "22-3" metal, has been demonstrated in a laboratory furnace at 1000 °C. For higher temperature experiments, sensor-input waveguide assemblies made entirely of Carpenter "22-3" metal will be employed. These assemblies, presently being fabricated, must be welded with an electron beam or electric arc to assure proper performance to 1400 °C operating temperature.

Success of the second microwave temperature sensor depends upon the emission of electromagnetic energy from a heated body. Ideally, the heated body is a black body for which the "apparent" and thermometric temperatures are equal. At microwave frequencies a matched load appears nearly as a black body. For initial experiments to 700 °C, a heated body of shaped carbon should provide a satisfactory impedance match. Calibrations and preliminary tests of the radiometer, which measured the amount of emitted signal, indicates that suitable sensitivity and response will be achieved for planned experiments.

Experiments will be performed with the radiometer to 1000 °C subsequent to development of a matched load operable at high temperature. Both circular and rectangular cross section waveguides will be used for signal transmission between sensor and measuring instrumentation.

Infrared Techniques

Most of the recent work on the infrared absorption hygrometer has concerned the optical and mechanical components. Past experience has shown that sensitivity is rapidly lost by peaking the absorption sectors of the rotating chopper-filter wheel too far on the short wavelength side of the 2.6-micron water vapor absorption band. On the other hand, if the absorption sectors are peaked on the long wavelength side, the instrument becomes too sensitive to carbon dioxide. While very good operation has been achieved, with adequate sensitivity to water vapor and no sensitivity to carbon dioxide, using chopper-filter wheels with absorption quadrants centered at 2.52 microns, it is believed that the optimum design will include a carbon dioxide cell in the radiation path to remove

any radiation characteristic of carbon dioxide. This will permit the use of absorption quadrants having peak transmission centered on the water vapor absorption peak.

The developmental rotating, balancing wedge filter holder appears to perform properly and permits the use of a wedge filter of much simpler design. A holder has been installed to support a wedge filter used in manual null adjustment or zeroing of the hygrometer. Considerable development remains to be done regarding the establishment of procedures for fabricating, in a reproducible manner, wedge filters with desirable characteristics, especially for the balancing wheel.

Upstream Boiling Burnout (D. R. Dickinson)

One objective of the Upstream Boiling Burnout Program is to develop a better understanding of boiling burnout in general by inferring a mechanism which could account for the occurrence of burnout in upstream locations. Work on such an interpretation of test results has continued.

At the conditions under which these tests were performed (mass velocity of 3 to 7 x 10⁶ lb/hr-ft², steam qualities at burnout greater than 10%, and a pressure of 1500 psig), an annular flow pattern probably existed in the test section. With this flow pattern, there is a film of liquid on the wall of the heated tube and a core of vapor with suspended water droplets in the center. When the tube is heated, water is evaporated from the surface film and burnout occurs when evaporation has proceeded to the point where the film disappears, and the heated surface is no longer wetted. In general, liquid water may be supplied to the heated surface at a given point by flow through the liquid film from the upstream end and/or by deposition of liquid drops. The following evidence suggests that deposition is the important mechanism under the conditions of these tests.

- Film flow would always result in burnout at the downstream end. Burnout at other locations (upstream burnout) requires sufficient deposition to rewet the surface and provide cooling downstream of the burnout point. To obtain upstream burnout in a uniformly heated tube as used in these tests, the deposition rate must be greater downstream than at the point of burnout.
- In several tests the heat flux was increased well beyond burnout, and burnout spread to cover a larger portion of the heated surface. The surface temperature was a continuous function of heat flux with no sudden jump in temperature at burnout. This implies that liquid continued to be supplied to the surface by deposition at a rate

corresponding to that at the burnout heat flux and that only the heat flux in excess of the burnout value was removed by convection to the steam. If the liquid had been supplied by film flow, the supply would have been sharply reduced to zero at burnout, all the heat would have to be transferred by convection, and there would have been a sharp temperature rise at burnout.

These tests were run at higher mass velocities than most burnout studies. At lower mass velocities there is less turbulence and more of the liquid will be in the surface film and less in suspended droplets. Thus, film flow may be relatively more important than droplet deposition in maintaining a wetted surface at lower mass velocities. This may be one significant difference between the conditions of these tests and other work where upstream burnout is not found. It could also explain why the burnout heat flux increased with increasing mass velocity in these tests, whereas more commonly at these qualities it is found to decrease. An increase in mass velocity will increase the droplet deposition rate by increasing turbulence and thus raise the burnout heat flux where deposition predominates. However, the film thickness will be reduced by increased mass velocity, and thus burnout heat flux will be reduced if liquid is supplied to the surface primarily by film flow rather than deposition.

As pointed out above, the occurrence of upstream burnout in a uniformly heated tube requires that the droplet deposition rate be higher downstream of burnout than at burnout. One possible mechanism which can be postulated to explain this is that the droplets suspended in the steam are reduced in size by turbulence as the mixture moves through the tube and that the smaller drops are more easily deposited.

The larger drops have larger inertia and thus are less apt to be moved radially by local steam turbulence. Since a higher mass velocity and steam quality will provide higher turbulence, these conditions would be expected to result in smaller droplets. If the above mechanism is correct, this could explain the exceptionally large increase in burnout heat flux with mass velocity and the increase in burnout heat flux with quality which was observed at the higher mass velocities.

UO₂-PuO₂ Fuel Cycles for Fast Reactors (E.A. Eschbach)

Effect of Operating Parameters on Breeder Reactor Doubling Time

Operating parameters representing both oxide and carbide fueled breeder reactors were selected and then varied to determine their impact on breeder reactor doubling time, which must be carefully defined.

Doubling time has been defined in many different ways. A comparison of doubling times from separate sources without a comparison of definitions is dangerous. Doubling time is defined here as the time required by a breeder to double the total of the amount of fissile material charged in the initial core, and the out-of-reactor inventory necessary to maintain reactor operation. Currently, the evaluation is simplified by only considering compound doubling times. Compound doubling assumes that there is a large system of like reactors and as fissile material becomes available for use it is placed in a reactor. The system must also provide available fissile material as required for the reactors without delay to reactor operation. Fissile material must, therefore, be fabricated into fuel elements, shipped to a reactor site and ready for insertion into a reactor before it is considered available.

The parameters selected as being representative of oxide and carbide fuel operation in a breeder reactor are presented in the following table. Calculations compute a 15.1 doubling time for an oxide fueled breeding reactor and an 8.1 doubling time for carbide fueled breeder.

1000 MWe Breeder Reactor

	<u>Oxide Fuel</u>	<u>Carbide Fuel</u>
Breeding Ratio	1.20	1.40
kg Fissile in Core	2,000	2,000
Number of Zones	3	3
Capacity Factor	0.8	0.8
Exposure MWd/MT Fissile	500,000	500,000
MWd/MT	100,000	100,000
Out-of-Reactor Time, Years	1.0	1.0
Compound-Interest Doubling Time, Years	15.11	8.13

The doubling time calculated above is based on the values selected for the parameters. The next step in evaluating breeder reactor performance is to determine the sensitivity of the doubling time to the variations in each parameter.

Out-of-Reactor Time. Out-of-reactor time is the time that fissile material spends outside the reactor core. The major contributors to out-of-reactor time are: spent fuel reprocessing, fuel element fabrication, transportation, and inventory. The out-of-reactor time has a significant impact on breeder reactor doubling time because increasing the out-of-reactor time increases the out-of-reactor inventory of fissile material that must be doubled.

Although the slopes of the oxide and carbide fuel doubling times are quite different, the percentage change in doubling times for an incremental change in out-of-reactor time is nearly the same. For example, changing the out-of-reactor time from one year to one-half year reduces the oxide fuel doubling time 22.8% and the carbide fuel doubling time 24.2%. In absolute terms, however, the oxide doubling time was reduced 3.4 years and the carbide just under two years.

If all out-of-reactor time could be eliminated, the oxide fueled breeder would double in 8.2 years and the carbide fueled breeders in 4.2 years.

Core Size. The decrease in doubling time due to reduction in core size is approximately the same for both oxide and carbide fuels even though the slopes of the curves are different. Improvements in doubling time can be obtained by increasing the specific power of the fuel. If the core size is reduced by one-half, the doubling time would decrease by about 25%. This would mean operating fuels to specific power levels of 600 watt/gram (most oxide fuels at present range between 150-340 w/g).

Zones. There was little effect observed on doubling time due to changes in the number of refueling zones. A slight improvement was made by going from 1 to 2 zones but after that the effect was extremely small.

Exposure. The effects on doubling time of fuel exposure on both carbide and oxide fuel are being computed.

Exposure Effects

Exposure was found to have a profound effect on the doubling time of breeder reactors. In the following simplified cases the breeding ratio was assumed to be constant over all the exposures considered (the actual effect would be a breeding ratio reduction as the exposures increase). Another factor not considered in these cases is the reduction in plant factor below the 80% used. (The plant factor would be reduced because of the increased downtime due to the increase in charging and discharging time as the exposure is reduced.)

The effect of exposure and out-of-reactor time on the doubling time for carbide and oxide fueled reactors has been examined. Where out-of-reactor time is zero, the doubling time increases as exposure increases. This is to be expected, since the lower exposures allow reintroduction of plutonium into the system faster and thus provide a shorter doubling time. A dramatic change occurs immediately after the out-of-reactor time increased from zero. The doubling time decreases as exposure increases until a minimum is reached at which time the doubling time again increases. The reasons for this reversal can be explained with the help of the table. At low exposures, neglecting the change in plant factor, the reactor is more efficient in plutonium production than at higher exposure. This is borne out in the zero out-of-reactor case. When an out-of-reactor time greater than zero is used in conjunction with a low exposure, a large out-of-reactor inventory is needed. For example, at 20,000 MWd/tonne fissile the out-of-reactor inventory is 3000 Kg, while the core itself requires only 2000 Kg. The total inventory being doubled is 5000 Kg. Compare this with an exposure of 500,000 MWd/tonne fissile. The total inventory is smaller than just the out-of-reactor inventory for 20,000 MWd/tonne fissile case. When the incremental decrease in out-of-reactor inventory becomes very small and when the plutonium reintroduction is delayed for long periods by the increased exposure of the fuel in-reactor, there can be an increased doubling time.

One Month Out-of-Reactor Time

<u>Exposure</u>	<u>In-Reactor Inventory (in Kg)</u>	<u>Out-of-Reactor Inventory (in Kg)</u>	<u>Total Inventory (in Kg)</u>
20,000	2,000	3,000	5,000
50,000	2,000	1,200	3,200
80,000	2,000	750	2,750
100,000	2,000	600	2,600
200,000	2,000	300	2,300
500,000	2,000	120	2,120
1,000,000	2,000	60	2,060
2,000,000	2,000	30	2,030
5,000,000	2,000	12	2,012

PLUTONIUM UTILIZATION PROGRAM (F. G. Dawson)Fuels DevelopmentHigh Exposure Plutonium Studies

Work has been initiated by Computer Sciences Corporation personnel to adapt a Lawrence Radiation Laboratory (LRL) computer program to the new 1108 machine. This program was delivered intact to us by LRL for our use in the plutonium program. Basically, the program locates photo peaks, computes the best fit after correcting for background and integrates the areas under each photo peak. While this will be a big help in data analysis, it is still not complete. The next step in making the program complete is to determine experimentally the over-all efficiency of our spectrometer system and then incorporate this function into the computer program. Once this is completed, we should be able to obtain yield data in addition to energy information.

Our existing computer plot program was modified to include provisions for plotting two independent sets of data after normalizing one set to the other. This feature allows us to make comparisons of energy spectrum changes as a function of time directly. In addition, it makes reproduction of the figures much easier by eliminating the need for hand plotting.

Underwater Profilometer

The underwater profilometer was delivered and assembled in the 308 Building for checkout and inspection of nonirradiated fuel rods. The instrument was checked by the seller's representatives and the performance was equal to or better than that specified. It was recognized by the seller's representative that slight improvements were desirable, and the components to accomplish this are being supplied. After inspection of the nonirradiated fuel rods has been completed, the profilometer will be installed in the PRTR adjacent to the FERTF fuel tray.

Project CAB-002

The vacuum fuel rod welding chamber and associated vacuum system have been accepted from the seller and were delivered on plant. Installation is scheduled to start during the last week of the month of June.

PRTR Testing

Irradiations are continuing in PRTR to determine the allowable reactor power level based on maximum fuel temperature considerations. PRTR power level is determined by the linear rod power that produces maximum permissible fuel temperatures just below melting.

The highest power rod examined was irradiated at 18.6 kW/ft. Results of the fuel structural analysis agree well with calculated values using a semi-empirically derived thermal conductivity curve for vibrationally compacted fuel that indicates fuel melting at 20.5 kW/ft. Results of the most recent analysis and previous examinations are summarized as follows:

Rod Power (kW/ft)	Approx. Time at Max. Power (hours)	Columnar Grain Growth Radius (%r)	Indicated Max. Fuel Temp. (°C) When Col. Grain Growth is Assumed to Occur at		Calc. Max. Fuel Temp. (°C)
			1800 °C	2000 °C	
16.8	48	50	2125	2460	2490
17.0	86	60	2300	2500	2500
17.9	80	60	2300	2500	2580
18.6	40	64	2400	2680	2650

The data illustrate that the columnar grain growth temperature is greater than 1800 °C for the irradiation times involved and that, as expected, structural equilibrium is attained more rapidly at the higher rod powers. Results of the examinations also illustrate the time dependency of structure formation.

The current irradiation is being conducted at a maximum rod power of 19.7 kW/ft which is 4% below the projected onset of melting and probably the limiting rod power for batch core operation. Estimated maximum fuel temperatures are 2740 °C.

Irradiation of the hot-pressed UO₂-PuO₂ 19-rod cluster pellet element is continuing satisfactorily in the FERTF. The maximum rod power is estimated to be 21.5 kW/ft with an associated maximum fuel temperature of 2600 °C.

Long term fretting and wear tests are continuing with a prototype 8-rod FERTF test element in the out-of-reactor EDEL test loop. The element has been exposed for approximately 45 operating days under anticipated FERTF coolant conditions. Periodic examination of the element has not revealed any noticeable fretting or wear condition between element components. A slight loosening of the two bottom circumferential strip bands occurred and is being evaluated during continued operation.

ETR Loop Testing

The PRTR prototype UO₂ defect test is operating satisfactorily in the ETR P-7 loop. The measured tube power is 305 kW at a reactor power of 175 MW versus a predicted tube power of 302 kW. If the peaking factors used in the predictions are correct, the maximum linear rod power is approximately 29 kW/ft with an estimated 63% of the radius molten at the plane of the defect.

Activity release to the coolant is comparable to previous defect tests and is causing no operational problems although there have been no power reductions up to the present time. Bursts occurred at the start of power level increases and rapidly attained an equilibrium level upon continued operation. One small burst occurred during steady-state full power operation. Barring any problems, the test will continue for approximately 20 days.

TREAT Transient Testing

Recent discussions with ANL indicate that a considerable amount of work remains to be done on the highly instrumented piston autoclave before it can be used in the proposed ANL-BNW joint program for transient testing thermal reactor oxide fuel rods. Design modifications, construction, and testing will take at least six months. In an effort to avoid a delay in the program, a simpler, less expensive autoclave design is being proposed by BNW for testing single fuel rods rather than three-rod clusters as originally planned.

A study to determine minimum fuel enrichment requirements in subsequent test rods was completed. The results of the study indicate that fuel rods enriched with 5 at.% U-235 (or equivalent Pu content) will provide the maximum energy levels desired and also provide a self-shielding that is not too different from that encountered in commercial power reactor fuels. Recent transient experiments with vibrationally compacted UO₂ (5.01 at.% U-235) fuel rods in the transparent autoclave have confirmed the results of the study. Agreement was reached between ANL and BNW to use fuel enriched with 5 at.% U-235 for the majority of subsequent tests.

Instrumented Fuel Rods

Four fuel rods instrumented to measure fuel rod gas pressure and temperature are operating satisfactorily in PRTR. The fuel rods have operated at reactor power levels to 64 MW and fuel temperatures of approximately 2700 °C without operational difficulties.

Though reactor operation has been limited and fuel rod pressure buildup has not been significant ($P_{\max} = 44$ psi; fuel exposure <4000 Mwd/tonne), fuel rod gas release seems to be following a definite trend. Fuel rod gas pressures increase to a maximum value in a stepwise manner during reactor startups, then level off or decrease slightly during steady-state operation. No gas release has been observed during reactor shutdowns.

It appears that at very low exposures most of the gas release in PRTR fuel elements occurs during startup.

Reactor Physics

D₂O Moderated Systems

Americium and Curium Production. Americium and curium isotopic concentration data from a highly exposed plutonium-aluminum sample irradiated in the PRTR has been received from the Analytical Chemistry Section. A preliminary analysis of this data shows that the results are of the expected order of magnitude. On this basis three more samples (with different exposures) have to be submitted for analysis. When the Am-243 and Cm-244 data are available from all of these samples, an attempt will be made to extract the Pu-242 and Am-243 neutron capture cross sections from the data by use of least squares methods.

Analysis of B-10 in D₂O. Measurements have been made in the Thermal Test Reactor (TTR) to determine the amount of B-10 in samples of D₂O taken from the moderator of the PRTR during the batch core experiments. Samples were taken each time changes were made in the boron concentrations of the D₂O moderator. Thirty-five samples were obtained from the critical tests and five more from the power tests to date. Analysis of the data from the TTR measurements is about 50% complete.

Batch Core Experiment in the PRTR. A paper entitled "Critical Experiments with the UO₂-2 wt% PuO₂ Batch Core in the PRTR," by R. I. Smith, J. W. Kutcher, and J. H. Lauby, was presented on June 12 at the 1967 Annual Meeting of the American Nuclear Society in San Diego, California.

A document entitled "Critical Experiments with Batch Core Fuel in the Plutonium Recycle Critical Facility," by J. W. Kutcher, J. H. Lauby, W. L. Purcell, L. C. Schmid, L. D. Williams, and J. R. Worden, has been written and is now in the publication stage.

Analysis of the batch core power tests in the PRTR is under way. These tests are still in progress but should be completed shortly.

Technical Assistance to PTU. Technical assistance is being furnished to the Process Technology Unit, PRTR Section, Engineering Services Department. Calculations have been performed to estimate the tube power in channel #1550, with a 19-rod UO₂-2 wt% PuO₂ cluster in H₂O, as a function of coolant temperature. A PRTR test to confirm these calculations is being planned.

RBU Monte Carlo Calculations. A paper entitled "Analysis of a UO₂ 19-Rod Cluster Experiment with the RBU Monte Carlo Code" was presented at the 1967 Annual Meeting of the American Nuclear Society at San Diego, California. On the same trip discussions were held with two of the formulators of RBU: J. R. Triplett, General Atomics, San Diego, California; and M. Temme, Lockheed, Palo Alto, California.

The subjects discussed included the origin of some of the ideas used in RBU and some of the problems associated with the use of RBU.

H₂O Moderated Systems

Gamma Scanning of EBWR Fuel. Seventeen rods removed from the EBWR in February 1967, are now available for gamma scanning. This work has begun, and gamma ray data from pin number EP98 have been taken. Pin EP98 was removed from location 36 of fuel element #46, which would locate this pin at the center of the reactor during its irradiation from November 1966 to February 1967. A preliminary plot of the data shows that the power distribution is skewed toward the lower half of the fuel rod, the peak being 11 inches from the bottom of the fuel. Very little burnup occurred in the top one-third of the rod.

Fabrication of PRCF Fuel Rods

About 800 (4 wt%) PuO₂-UO₂ test fuel rods are needed for critical experiments in the PRCF. Because commercial procurement of rods was not possible to meet FY-1967 programmatic needs of the program, approximately 280 rods were fabricated internally. Commercial procurement to obtain the balance of the rods during FY-1968 is now being completed under the two-step procurement procedure.

About 14 of the total 58 kilograms of uniform isotopic plutonium nitrate supply previously obtained was processed to the metal, vacuum strip-cast, low temperature oxidized, and calcined to yield a 11.4 kg PuO₂ supply. The purity and isotopic content of the PuO₂ were determined. Of particular interest is the isotopic content which is as follows: Pu-238 = 0.27, Pu-239 = 75.4, Pu-240 = 18.2, Pu-241 = 5.16, and Pu-242 = 1.17.

This PuO₂ was blended with natural isotopic UO₂, pneumatically impacted, crushed, sieved, blended for particle composition, and vibrationally compacted. Rods were then decontaminated, welded, tested for homogeneity, and leak-checked before storage. Fabrication of the 280 fuel rods is to be completed by June 30.

Seven possible commercial fabricators were informed of the proposed work. Information submitted included the test fuel rod design drawings, specifications of fuel, and instructions about the two-step procedural requirements. Under step one, which is now in progress with commercial vendors, technical proposals are being considered. Five of the possible seven commercial fabricators responded with technical proposals. Areas of variance in these proposals are being resolved with the companies. Request for pricing of fuel rods is planned with notification to offerors under step two planned for release about July 15. About 200 test rods are being requested for delivery on November 1, 1967; other options are included about possible delivery date for additional fuel rods.

Heterogeneous Experiments

A joint program between the CNEN and the AEC will study power sharing and power peaking in heterogeneous loadings in the PRCF. Design of experimental hardware is complete. Shop work will begin immediately. Modifications are also in progress for UO_2 fuel and fuel followers to be used in the experiment. Completion goal for the entire work is about September 1.

PuO_2 Particle Size Studies

Modifications to the induction plasma arc equipment for spheroidizing PuO_2 were completed. Preliminary tests indicated a significant improvement in plasma arc stability. Work included a Freon-cooled canister (enclosing the plasma gun) and a Freon-cooled collecting unit. The unit was also modified to provide sufficient gas flow to prevent arc-flash ground previously experienced in the prototypic unit.

A set of fuel rods containing solid solution UO_2 -2 wt% PuO_2 (24% Pu-240) was completed. The set consisted of four 20-inch fuel columns and two 10-inch fuel column rods clad in Zircaloy.

A series of rods containing PuO_2 spheres from the pneumatic-impaction and jetmilling sequence has been tested in the PCTR to determine the reactivity effect in graphite moderated lattices caused by increasing the PuO_2 particle size through the range of 0, 100, 200, and 350 microns. The fuel contains 2.0 wt% PuO_2 in PuO_2 - UO_2 with a nominal 8 wt% Pu-240 and natural uranium.

The first experiment contained 0.5 inch diameter fuel in a 4.0-inch square graphite cell. Foil activation measurements were made to determine the relative reaction rates of the cell components. Reactivity measurements indicate a general decrease in k_∞ with increasing particle size. Analysis of the foil measurements is under way to calculate the magnitude of the decrease in k_∞ .

Code Development*

BNW Master Library. The BNW Master Neutron Cross Section Library was updated to include additional and more accurate data. The following isotopes were updated in June:

Sodium-23	Indium-115
Iron-54	Thulium-169
Iron-56	Hafnium-177
Iron-57	Hafnium-179
Iron-58	Plutonium-242.

All of the changes were made because of newer resonance parameter information.

*Partially supported by 02 Program funds.

Subroutine BCDRD. BCDRD was modified to improve the BCD to binary conversion scheme and to bypass the executive system double precision interpretive routine. This modification was necessary because the former version of BCDRD would not work with the 1108 double precision hardware when it replaces the interpretive routine.

A comparison of the conversion time requirements of the modified BCDRD on the Univac 1107 and 1108 was made. The average amount of time required to read a card dropped from 3.3 to 2.44 ms on the 1108. The average amount of time required to convert a number on a card into binary form dropped from 2.52 ms to 0.42 ms on the 1108. The comparison involved 605 cards and 3216 data words.

ENDF/B Library. Legendre scattering coefficients were transformed from the center of mass system to the lab system and punched in ENDF/B format for the following isotopes: Lu-175, Au-79, Dy-164, Eu-151, Sm-149, Lu-177, and Eu-153. Data for deuterium could not be punched because code characteristics led to an overflow condition in the transformation matrix when the ratio of the neutron mass to the mass of the nucleus was greater than 0.5.

BNW Master Library. The program Unicorn was used to calculate low energy cross sections from resonance parameters for Pu-242 and Tm-169. The resonance parameters and cross sections were punched in BNW Master Library format.

Program UPDATE. The computer program UPDATE, which is used to update the BNW Master Library, was finally converted from the Univac 1107 to the 1108. Conversion was delayed until the system interpretive routine that performs the double precision arithmetic was converted to the Univac 1108. The card punch option is very time consuming in the 1108 version, at times requiring on the order of 0.1 second per card. This problem should disappear when the system has a sufficient number of drum buffers.

BARNS-II. Several revisions were made to BARNS-II to correct some errors and to rearrange some computing algorithms to reduce the likelihood of system interrupts. The code was also used several times in debugging other codes derived from BAPNS-II when they were changed to reflect recent BARNS-II revisions.

HRG Data Tape. The latest HRG data tape is HRG 060767. For this tape the data for H₂O and D₂O have been corrected to represent value per molecule, rather than value per atom, as had inadvertently been given originally. The nuclide numbers for the Westcott and Leonard versions of U-235 and Pu-239 data have been restored to their values prior to HRG 020667. The latest tape contains data for 240 nuclides.

GASKET. The computer program GASKET⁽¹⁾ has been compiled and is running on the Univac 1108. This code generates the scattering law $S(\alpha, \beta)$ for thermal neutrons for all common moderators.

Reactor Engineering Development

Pu Optimization Studies

A rough draft of a report describing the design code, REPP, for BWR's and PWR's has been completed and will be submitted to Technical Publications for issuance shortly.

A change in plans for development of a detailed design code for BWR's and PWR's was made. Instead of modifying the present survey code, FULCYC, it is now intended that the design code, REPP, be added to the analysis code, BOLERO, along with an economics package. Required computer programming is currently under way.

PRTR Fuel Element Rupture Test Facility

Rupture Loop Particle Separator. A clear plastic section is being attached to the flow separator downstream of the swirler. This section will be used to obtain the swirl angle by use of a high speed camera. Testing is scheduled to start by June 27.

The special equipment, which will be used to obtain a more accurate particle size distribution, has arrived onsite and is currently being prepared for use.

Fuel Element Testing. Testing of the FERTF basket-type element continued during the month. To date, the element has been tested a total of 1078 hours under the following conditions.

<u>Temp.</u> <u>OF</u>	<u>Pressure</u> <u>psi</u>	<u>Flow</u> <u>gpm</u>	<u>Pump Speed</u> <u>rpm</u>	<u>pH</u>	<u>Total Time</u> <u>hrs</u>
475	1080	120	1700	7-9	300
475	1050	110	1500	7-9	601
475	1050	70	1010	7-9	177

On June 10, the element was removed from the pressure tube for examination. Concurrently the tube was examined. The depth of scratches and fretting marks was measured with a modified dial indicator. None of the scratches were more than 0.002 inch deep. Measured depth of the fretting marks was 0.0015 inch.

(1) J. V. Koppel, J. R. Triplett, Y. D. Nalibobb, GASKET: A Unified Code for Thermal Neutron Scattering, GA-7417, General Dynamics Corp., General Atomics Division, San Diego, Calif. September 1966.

An attempt was again made to photograph selected portions of the tube interior. This was not completely satisfactory; however, some usable information was gained and will be factored into a third attempt.

The operating spare seals and bearings for the EDEL-1 pump have been received. The pump has operated 1820 hours since its last overhaul and shows no sign of pending failure. The previous operating time between pump overhauls was 3200 hours.

Materials Development

Aluminum Corrosion in Borated D₂O

Aluminum coupons suspended in the moderator system of PRTR in contact with D₂O containing boric acid (20 ppm B-10) or in contact with the helium gas blanket were found to be uniform in appearance with no pits or unusual discoloration. Many of these coupons had been exposed 701 days, during which the reactor was in operation a few months.

The 35 coupons removed showed weight gains varying slightly with the type of aluminum alloy composition. Average weight gains in mg/dm² were 60 mg for 5050 alloy, 61 mg for 5052 alloy, 73 mg for 6063 alloy, and 85 mg for 6061 alloy. The greatest weight gain was for a 6061 specimen (110 mg/dm²) exposed 701 days in the lowest position in the moderator where it was continuously submerged.

Three coupons representing the top, middle, and bottom regions of the moderator were stripped. On the basis of metal loss, weight gains lay between the values for the trihydrate and monohydrate oxides indicating small loss of material from the aluminum surfaces.

In general, appearance weight gain and metal loss agree with previous test experience and do not reveal any unusual corrosion of aluminum in the moderator system.

PRTR Pressure Tube Evaluation

The PRTR reactor safeguards program includes study by destructive examination of the effect of reactor environment upon the Zircaloy-2 pressure tubes. Work in progress is composed of determining a hydrogen limit on unirradiated tube specimens of different flaw length and crack propagation tests at various temperatures. This work is continuously improving the basis for judgment of irradiated specimens.

A crack propagation test was run on a 20-inch specimen of annealed PRTR tubing with a 1½ inch slot milled 80% through its maximum wall thickness. The test was run at 150 °C and burst at

a pressure of 3350 psi. Using Lamé's equation, a hoop stress at failure of 38,500 psi was calculated. This test completed the hoop strength versus temperature curve for a $1\frac{1}{2}$ inch flaw length out to a temperature of 300 °C. A slope of -10 psi/°C was attained.

The effect of hydrogen on fracture surface appearance of a broken tube is being studied; three broken tubes that were identical in all but their hydrogen content were selected for this study. The three tubes all had $1\frac{1}{2}$ inch slots milled 80% through their maximum wall thickness and were pressurized to failure at room temperature. The "as received" tube, the one containing 150 ppm hydrogen and the one containing 275 ppm hydrogen failed at hoop stresses of 34,000, 35,000, and 37,000 psi, respectively.

The three tubes have been sectioned and prepared for fractography. The replicas for fractography were taken from the web, $\frac{1}{4}$ inch from the end of the web, and 3 inches from the end of the web. These replicas will be examined by electron microscopy to see what effect the hydrogen content had on the fracture surface topography.

A tentative specification for defect detection limits for inspection of PRTR process tubes was prepared and distributed June 14, 1967.

Based on an overall safety factor of four, electronic inspection of PRTR process tubes must be able to detect:

- (a) with regard to potential catastrophic failure, a crack defect or flaw one inch long by $1/8$ inch deep;
- (b) with regard to potential leakage of coolant from the process tube, a crack defect or flaw $\frac{1}{4}$ inch long by $1/8$ inch deep.

Until adequate experience is gained with interpretation of signals from nondestructive electronic inspections, it is suggested that visual inspections of indicated flaws or defects also be made.

When inspections signal the presence of flaws or defects with dimensions half of those specified above, increased surveillance should be initiated to ascertain rates of approach to service limits. Reactor operation, just prior to detection of flaws $1/16$ inch deep or more, should be critically reviewed in an effort to ascertain cause so that if and when appropriate, operating conditions may be altered to halt or slow down the approach to service limits.

The present review of surveillance data suggests that investigations of the effects of hydriding, irradiation, and the combination of hydriding and irradiation on fatigue crack initiation, growth, and propagation would be most desirable.

Cycle Analysis

Pencil Lead Fuels

Seventeen RBU Monte Carlo problems have been submitted and the calculations on 12 have been completed, but analysis of the results has not been started. This study is using BNWL's latest cross section data and a most recent version of Monte Carlo which produces an auxiliary output tape for statistical analysis of the results. Two minor changes were made for this study to improve the speed for these small cells. The random log routine which is used to calculate the distance to collision was revised to use only one random number and to interpolate using integer arithmetic. The former method always used six significant figures in the interpolation while this method uses multiples of 2.38×10^{-7} mean free paths which is less than six significant figures for those cases (1 out of 16) when the distance to collision is less than 0.0645 mean free paths. It will be less than three significant figures in one of 32,000 cases. Furthermore, in one out of 8.4×10^6 cases, it will return zero as the distance to collision rather than the very small number previously calculated.

The second change involved the flux tallies which previously were followed to 99.9% with the last 0.1% estimated by random choice. In this study the first 93.75% is tallied directly and remaining 6.25% estimated by random choice.

The first two cases repeated the uniform and pencil cases at 2% plutonia enrichment in natural urania. The other 15 are 5 variations of plutonia particle size at enrichments of 1.024%, 2%, and 3.9%. The nominal particle sizes were 512, 640, 800, 1000, and 1562.5 microns diameters. The particles were spheroids with axes 1.732 times nominal, nominal, and 0.577 times nominal. Thus the major axis was three times the minor axis. There are six possible orientations and approximately equal numbers were located with long, nominal, and short axes in each of the x, y, and z directions for each case. An attempt was made to locate the particles uniformly in both x, y, z, geometry and R θ geometry

Test Reactor Operation

Operating Experience

Pertinent data for the month are as follows:

1. Production	608.34 MWe
2. Hours Critical	317.9 hrs
3. Critical Efficiency	44.2%
4. Total Experimental Time Efficiency	46.3%

5. D₂O Losses

- a. Indicated Stack Loss (5/26-6/25) 1083 lbs
- b. Physical Inventory (5/26-6/23) 1607 lbs

6. Helium Loss 101,253 scf

The PRTR operated at a maximum power level of 64 MW this month. The reactor power level was limited by PRTR Test No. 136, Supplement 2, to a specific rod power of 19.7 kw/ft in the average Ring One fuel element. Total production, since the start of the batch core experiment, is 1362 Mwd's, or about 7% of the batch core goal exposure. The 55 fuel element batch core has accumulated 1162 Mwd's.

PRTR operation was interrupted by 15 shutdowns. Two were scrams caused by flow monitor trips, four scrams were caused by four separate events, one planned shutdown was made for liquid shim adjustment, and eight were intentional shutdowns to correct operating conditions.

There were 11 major outages as follows:

Date	Reason for Shutdown
May 29	Shut down 46.1 hrs to locate and correct the cause of high D ₂ O losses.
June 1	Shut down 21.4 hrs to locate and correct the cause of high D ₂ O losses.
June 2	Scrammed by pressurizer low pressure. The scram occurred during reactor heatup and is believed to have resulted from instability in the secondary coolant system. After the scram, steam was observed spraying from a crack in the elbow of one of the three vent lines on the primary heat exchanger side of the primary heat exchanger, HX-1. The elbow was removed, examined, and found to be pitted and cracked. The other two elbows were removed after radiography revealed they were also in poor condition. The heat exchanger and remaining associated piping received extensive examination. The heat exchanger shell was examined visually, while the piping inspection utilized radiographic techniques. No further problem areas were located. A third party inspector approved the vent line elbow repairs. The outage required 178.4 hrs.
June 10	Scrammed by a FERTF high pressure fluctuation. The outage required 8 hrs.
June 10	Shut down for 33.6 hrs to correct high oxygen in the FERTF coolant, and to correct the earlier FERTF pressure oscillation problem.

- June 12 Scrammed by reactor period. The outage required 6.2 hrs.
- June 14 Shut down for 6 hrs to correct high oxygen content of the FERTF coolant.
- June 18 Shut down for 16.3 hrs because of high collection rates and low isotopic purities in the D₂O recovery system. Seven leaking nozzle caps were found to be responsible for the high D₂O collection rate, while the degradation was traced to a leaking jumper hose on the rotating shield cooling line.
- June 20 Shut down for 6.2 hrs to correct high oxygen in the FERTF coolant.
- June 21 Shut down for 18.1 hrs to correct high oxygen in the FERTF coolant.
- June 22 Shut down for 74.9 hrs because the turbidity of the filtered water exceeded the operating limits. An alternate water supply was valved in while the turbid water condition was corrected. The outage was extended because the FERTF cleanup ion exchangers, RL IX-1 and 2, broke through and, in the process of valving the flow to bypass these ion exchangers, RL-14 was opened and resin back-washed from RL IX-1 into the FERTF coolant. Process channel 1550 was disconnected from the FERTF and connected to the primary coolant system to facilitate cleaning the resin from the FERTF.

During the first part of the month high heavy water losses continued to be a problem. The reactor was shut down twice to locate and correct a leak which was found to be at the ring seal flange joint at the base of H-95. After repairing the leak, heavy water losses averaged 11 pounds per day during the succeeding week of reactor operation.

The pressurizer safety relief valve, H-95, started leaking during reactor operation. Because of its physical location in relation to the ring header vent valve, P-12, about 125 pounds of primary coolant were lost before the leak source was located. The reactor did not scram nor was there any unusual variation in pressurizer pressure or level. This is the second time in the past two months that H-95 has started leaking while the reactor was operating. Bench tests proved that the valve was unsatisfactory since it leaked helium at 850 psig while it held water at 1150 psig.

During these major outages, the general program of reducing D₂O leaks continued. Two more angle valve leakage collection lines

were disconnected, the valve bodies plugged, the leak collection lines capped, and the valve packings tightened. There are now 32 angle valves that have been plugged to control packing leakage.

"Crud" levels continued to be low in the primary coolant system under pH 7 operation at power levels to 64 MW. The examination of irradiated fuel elements showed that there were small areas near the ends of the fuel elements and on the fuel element bands with thin layers of material that is dark reddish-violet in color.

Variations of flow through the primary system filters have suggested that reversible changes in the crystal structure and density of the crud will occur as a result of small changes in the primary system dissolved oxygen concentration.

Borescope examination of the internal surfaces of the calandria and shim wells showed that these surfaces were in good condition. Some material was visible in the bottom of the shim wells, and some flaking rust was visible in the carbon steel top shield penetration. Samples of solids from the moderator side leg filter cartridges were removed and were being analyzed at month end.

Seventeen process tubes were examined. There were no significant changes observed since the previous examinations. Eighteen fuel elements were discharged from the reactor and inspected in the storage basin. All were in satisfactory condition for recharging into the reactor.

Process Technology

The two fission gas pressure measuring fuel elements (PRTR Test No. 124) continue to perform satisfactorily. Thirty pressure and temperature measurements were made on each of the four test rods. The maximum temperature and pressure observed were 518 °F and 53.5 psig in rod eight, fuel element 6520, in channel 1548.

The test of the pelleted fuel element (FERTF-22) was interrupted on June 24, when ion exchange resin was released to the FERTF due to backflow through a cleanup ion exchanger during a reactor outage period. A fuel rod from this element and also from adjacent fuel elements were removed for destructive examination. Irradiations in the FERTF will be resumed after cleanup of the loop.

The irradiation to determine the maximum allowable fuel element rod power continued (PRTR Test No. 136). This month, irradiations at a maximum rod specific power of 18.6 and 19.7 kw/ft were completed. Fuel rods were removed for destructive examination.

Considerable difficulties were experienced in controlling the FERTF oxygen concentration during the latter part of the month. Investigation to date has suggested that the increase in dissolved

oxygen was due to radiolytic decomposition during reactor operation without the recombination (or back-reaction) rate expected due to high temperature operation of the loop. FERTF Test 23 was prepared to continue this investigation.

The moderator corrosion coupon holder was discharged on 6/8/67, and the sixth partial coupon replacement was completed. Thirty-five coupons were removed, and 24 new coupons were reinstalled.

A corrosion coupon assembly was removed from the inside of the primary heat exchanger (HX-1) shell. One coupon was removed for detailed examination. The remaining assembly was examined visually, a new coupon installed, and the assembly reinserted into the HX-1 shell.

Improvement Work Status

Work Physically Completed.

NPC-407 - New A- and B-Cell Sump Pumps. As a part of Project BCP-007, new sump pumps have been installed in the process and experimental cells to improve recovery and control in the event of a flooding incident. The new 150 gpm capacity pumps are submersible and connected to emergency power. Original pumps were 100 gpm, non-submersible, and were on normal power.

NPC-433 - Removal of A-Cell Controls for Primary Pumps. Key-lock start-stop switches have been removed from the vicinity of the primary pumps. Experience showed they were not needed but were a source of malfunction in the pump controls.

NPC-405 - New Valve Guards for Water Chillers. Sintered metal guards for compressor valves have proven to be a very costly maintenance item because of frequent breakage and subsequent damage to compressors. They have been replaced by wrought metal guards.

Work Partially Completed.

Safety and Containment System Modernization. The safety and containment systems at PRTR are being modernized by installation of a second safety circuit and redundant containment and fog spray valves. About 75% of the safety circuit material is on hand, and detail design is 95% complete. Design is complete on dual containment valves. A minor amount of pre-outage work is in progress on all three systems.

Projects - Technical Guidance

BCP-007 - PRTR Waste Handling. This project is to provide improved waste handling capability and control of reactor systems during accident situations. The status of remaining work items is as follows:

1. New A & B Cell Sump Pumps (NPC-407). Installation is complete. Sealing of watertight electrical fittings has been done. Minor finish details remain.
7. Remote Venting of Primary Pumps (DC-291). The installation was completed previously, but faulty valves will require additional work. The manufacturer is being requested to provide improved replacement parts.
11. Rupture Loop Bypass Piping (DC-RL-22). Work remains at 98%. A chain operator for a valve was received but is not yet installed.

All other items are complete. Over-all completion of BCP-007 is estimated at 99%.

BCP-013 - PRTR Contamination and Waste Control. A revised Bacon-Davis Act work review was submitted to the AEC for approval. This revised review assigned schedule and hazards sensitive work items to Plant Forces. The status of each item on this project is as follows:

1. Replace C Cell Sump with a 100 gpm submersible unit. Design is complete.
2. Emergency pumping capability for transfer of D₂O from TKA-2 to DT-2. Design is complete.
3. Charcoal filters for recirculation units R-1, R-2, and R-6. Design is complete.
4. Activated charcoal filter for fueling vehicle. Redesign of this unit to incorporate comments has been started.
5. Activity monitor at manhole #3. Design change has been approved and issued.
6. Air sampling station. Drawings transmitted for approval.
7. Level indication for A, B, and C Cell Sumps. Design is completed.
8. Pumpout station at manhole #2. Design is complete.

AEC 193 - Fire Protection Improvements. This project is to provide sprinkler protection in the 309 Building in all areas except the control room and containment vessel. The complete project includes the 326, 327, and 306 buildings.

Preliminary drawings by the contractor were received during the month and comments returned. Construction work is expected to start in July or August 1967.

Instrumented Fuel Element Installation. Equipment will be installed in C Cell to monitor pressure developed in a fuel element in-reactor. No progress during June.

High Resistance Neutral Grounding - 2400 Volt System. The existing 2400-volt electrical power systems in the 300 Area operate as ungrounded delta. It has been agreed that these systems should be grounded, including the system at Building 309, to improve operating characteristics of the system by high resistance neutral grounding transformers.

One meeting and several discussions have been held during May and June regarding installation at Building 309. The concrete pad for this equipment has been installed. The electrical equipment is on order but has not been delivered.

NUCLEAR SAFETY

Containment Systems Experiment (J. M. Batch)

Containment Leakage Studies - Task A

Effect of Temperature Sampling Errors in Leakage Rate Measurements. A serious source of error in determining the leakage rate of containment vessels is the use of inaccurate average containment air temperatures. Such inaccuracies could result from inadequate sampling of the air volume within the vessel. During the series of leakage rate tests on the CSE containment vessel, air temperatures were measured by 19 platinum resistance temperature detectors (RTD's) distributed to provide one RTD for each 1600 ft³ of containment vessel volume. Leakage rate calculations were performed using the absolute pressure-temperature method and were based on changes of the volume-weighted average temperature indicated by all 19 RTD's.

During the month additional calculations were started to assess the probable magnitude of the error in the average containment air temperature resulting from use of both smaller numbers of RTD's and of disadvantageously located RTD's. Vessel leakage rates calculated from these less accurate average temperatures will be compared with those based on the average of all 19 RTD's. The calculations are continuing, and no conclusions are available at report time.

Fission Product Transport Behavior - Task B

Aerosol Transport in the Drywell Vessel. Large-scale experiments are planned in the CSE containment vessels to study the behavior and transport of fission products and their removal by both natural processes and engineered safeguard systems. Knowledge of

the concentration of airborne fission products as a function of time is important in the calculation of the consequences of major accidents and the determination of the acceptability of the proposed reactor site.

The first shakedown aerosol transport test (Run #D2.1) was performed in the CSE drywell vessel. Primary objectives of this test were the evaluation of equipment and techniques to be used in scheduled CSE containment vessel tests and to obtain mass transport data in the drywell vessel which could be used in mathematical models developed from ADF test data. Scaleup from ADF runs was by a volume factor of 65.

The drywell has a volume of 65 m^3 and an area-to-volume ratio of 1.54 m^{-1} . All surfaces are painted with Phenoline 302. The test was performed at isothermal and isobaric conditions (250°F and 49 psig). The molar steam-air ratio was 1.68. An iodine and cesium aerosol was rapidly injected by means of a steam jet injector using superheated steam as the motive fluid. Makeup steam replaced condensation and provided mixing for the vessel atmosphere. Seven sample clusters, each containing 12 Maypacks were provided to sample airborne aerosol concentration radially and axially within the vessel. Deposition coupons of bare carbon steel, Phenoline 302 coated carbon steel, stainless steel, and silver-plated copper were attached to cluster assemblies for measurements of integrated deposition on non-condensing surfaces. Ten temperature sensors and nine atmosphere velocity sensors were positioned to monitor radial and axial distributions of these variables. Samples of condensate were taken periodically during the 23-hour test duration.

Analysis of data obtained during the test is in progress; however, some preliminary results can be reported. Gas-phase concentration decreased exponentially with a half-life of $8 \pm 0.5 \text{ min}$ compared with 12 min predicted by the ADF model. The ADF model, however, used a correlation to estimate gas convection velocity obtained from steam condensation rate data. When actual measured velocities from the drywell test are used, the theoretical half-life of aerosol in containment will probably be in closer agreement with the observed value.

Aerosol mixing within the drywell as indicated by the seven radially and vertically spaced aerosol samplers was good. The standard deviations of the seven aerosol concentration locations measured at 13 min after injection were $\pm 15\%$ for iodine and $\pm 25\%$ for cesium. Sampling and analytical error is estimated to be $\pm 15\%$. Total airborne iodine concentration decreased from $660 \text{ } \mu\text{g}/\text{m}^3$ to $3 \text{ } \mu\text{g}/\text{m}^3$ in the first hour of containment and had decreased to less than $0.2 \text{ } \mu\text{g}/\text{m}^3$ after 23 hours and termination of the test. Cesium airborne concentration dropped exponentially more than three orders of magnitude in the first 90 minutes of containment, after which some change of slope was observed. Analytical uncertainties and

cross contamination problems became appreciable for cesium analyses beyond 90 minutes in containment and tailing of the exponential curve at later times may be due to these effects. The material balance at run termination showed about 25% of the iodine and less than 1% of the cesium remaining on the vessel walls. Material balances for iodine and cesium following steam decontamination were 78 and 104%, respectively.

Some 660 individual samples were analyzed for cesium and iodine activity in the course of the drywell run. Less than four calendar days were required to analyze and output the analytical data in tabled forms.

In summary, all run objectives were fulfilled. Performance of aerosol generation, injection and sampling equipment was basically satisfactory. Several relatively minor shortcomings were observed where improvements will be made. Similarly, operating techniques were adequate, and control of the run was satisfactory for an initial test. Aerosol behavior was basically as predicted by the transport model developed from the smaller-scale tests in the ADF. The shorter than predicted airborne half-life is believed to be the result of the convection currents within the vessel. Indicated velocities were about 600 fpm upward on the vessel axis and about 150 fpm downward at 6 inches from the wall at mid-height. These values will require correction to the atmosphere conditions of the test, and the appropriate correction factors are not yet available. It was observed that the leakage rate from the drywell increased very markedly at test conditions as compared to the pre-test leakage rates using ambient temperature air. Based on makeup air supplied to the vessel to maintain steady containment temperature and pressure, it was estimated that leakage rate increased by more than a factor of 10. The principal leak points were at the butterfly-type valves used in the vessel ventilation system.

Coolant Blowdown Studies - Task C

Blowdown Experiments. A cold blowdown test was performed in which nitrogen pressure was used to expel water from the reactor simulator vessel through a 5.189-inch ID orifice. The purpose of this test was to evaluate instrumentation performance during a short duration blowdown under conditions for which results can be accurately predicted. The initial pressure in the vessel before the blowdown was 1550 psig. About 100 ft³ of water was discharged in 3 seconds. The observed pressures and outflow rates during the blowdown were within 5% of calculated values. The load cell water weight measurements agreed well with the liquid level probe measurements even though the initial acceleration of the vessel caused the loadcell readout to swing off-scale.

The results of this run, together with those of the preceding three low temperature, gas-driven blowdowns, have shown that the performance of instrumentation for liquid weight, liquid level,

vessel and nozzle pressures, and vessel reaction force is suitable for the expected range of transient inputs in forthcoming hot blowdowns. The neutron densitometer also was to be used on this run; however, high voltage corona in the transformers precluded its use. Certain design deficiencies in the transformer must be corrected before the densitometer can be used. A paddle wheel vorticity indicating device was placed at the entrance to the outlet nozzle in this test. This device was installed to attempt a test of the hypothesis that a hollow vortex was generated which allowed nitrogen gas venting to begin while the liquid level was still above the outlet nozzle. The device was unfortunately destroyed by reaction forces during the blowdown, and no data were obtained.

This test concluded the planned group of cold water blowdowns. A series of hot blowdown runs will now be started to investigate blowdown of the reactor simulator vessel at representative PWR and BWR coolant conditions.

Radioactive Waste Solidification (A. M. Platt)

Phosphate Glass Process Chemistry

Sodium is added to the feed solution when PW-1 is processed in the BNL phosphate glass process to lower the viscosity of the evaporator solution and to lower the operating temperature required in the continuous melter. A series of measurements were made on simulated evaporator solutions at b.p. = 140 and 150 °C in which the additive Na concentration was varied incrementally from 1.12 to 2.42 moles per liter PW-1 at 100 gal/tonne. The major effect of adding sodium was to gradually change the homogeneous evaporator slurry, or suspension, to a nonhomogeneous slurry containing a heavier component, which settled out fairly rapidly at the boiling point. The reduction in viscosity which occurred as additional Na was introduced is probably connected with the changing characteristics of the solids in the slurry. The experimental results indicated that, in order to avoid macro formation of the heavier solids in the evaporator, the additive sodium concentration must be maintained at less than 2.0M, or the temperature must not exceed 140 °C.

The corrosion rates of 304L stainless steel and A55 titanium in the simulated evaporator solutions did not change significantly over the sodium concentrations studied. The corrosion rates measured for titanium were a factor of 4 to 10 below those of stainless steel.

Sulfate is also known to decrease viscosity in the evaporator, and sulfate concentrations from 0 to 0.6M were investigated with PW-1 phosphate glass solutions also containing 1.98M additive Na. The maximum decrease in viscosity which was obtained at b.p. =

140 and 150 °C was about 40%. The presence of up to 0.6M SO_4 in the feed did not affect the corrosion rates of 304L s.s., or A55Ti at simulated evaporator conditions.

A pronounced decrease in the corrosivity of PW-1 waste solution of titanium was achieved by the addition of aluminum nitrate. The addition of 0.4 mole of ANN to one liter of PW-1 solution (Meq/P = 0.9) reduced the titanium corrosion rate to less than one mil/month. This effect is being investigated further.

Spray Calciner Development

To evaluate the performance of an internal mix spray nozzle when using steam as the atomizing medium, an 8-hr run (DSC-5) was carried out on a PW-2 flowsheet; the calcine was collected in a drum. The nozzle operation during the run was very erratic with considerable nozzle pulsation being experienced. A pulse consisted of a sudden decrease in the feed side nozzle pressure and a resulting increase in the feed flow with the pressure and flow being just as quickly restored. It is considered likely that the pulsing resulted from condensation of the steam in the nozzle since no detectable change in the steam supply pressure to the nozzle was noted. It was also noted that the pulsations were greatly diminished when the steam temperature at the nozzle approached 300 °C, and the feed flow did not exceed approximately 12 liters/hr. After the run some deposition was observed on the calciner walls which was not unexpected considering the erratic operation. The atomizing gas preheating system has since been modified to increase the steam superheat at the nozzle and efforts to improve the reliability of steam as an atomizing medium will be continued.

During the month a 24-hr run (DSC-6) was carried out with the calcine being melted directly in the receiver pot. The run was carried out on a PW-2 flowsheet using an internal mix spray nozzle and air atomization. Solid NaPO_3 was added directly to the receiver pot via the WSEP vibrating trough solids feeder.

A total of 334 liters of feed (454 liters/tonne) was consumed during the run at rates ranging from 11 to 16 liters/hr. The atomization of the feed was adequate as no gross deposition occurred in the calciner. A total of 35.7 kg of 3-8 mesh NaPO_3 was added to the receiver pot during the run via the vibrating trough solids feeder. The feeder's performance was quite satisfactory with no bridging or plugging problems experienced. The reduction of hopper diameter to 9½ inches permitted an evaluation of the hopper discharge rates in sufficient time to enable the taking of appropriate control to meet the needs of the process. The estimate of the total solids added during the run as indicated by the change in hopper level was within 3% of that required by the process and within 4% of that actually added as determined by the weight of the hopper contents both before and after the run.

The in-pot melting proceeded smoothly until the last 3 hours of the run at which time the melt level became covered with calcine and the run was stopped. The pot was filled to within 18 inches of the top with 132.5 kg of product being collected. The product level was within 6 inches of the top of the furnace, and it became apparent that with this condition it was not possible to maintain the calcine at a sufficiently high temperature; consequently, the last portion of the calcine was only partially sintered. The feed to product volume ratio for the run was 6.4.

During the month the occurrence of erosion on the inside of the internal mix nozzles was observed. The erosion was noted on the internal surfaces of the nozzle tip immediately beyond the point where the feed is injected into the atomizing gas. As shown in the table, the erosion was confined (with one exception) to the inside of the nozzle tips and based upon preliminary observations it had little effect upon atomization in those cases wherein it was confined to the inside of the tip. The enlargement of orifice on tip #2 changed the flow characteristics considerably and may have affected atomization to the extent that a higher mass ratio of air to feed was required to provide the same degree of atomization as attained previously. The condition was easily noted on the calciner since at a given atomizing gas pressure and liquid flowrate the gas consumption of the nozzle had increased considerably and the liquid side pressure had decreased as a result of the increased flow area.

Nozzle Tip Erosion Data

Tip No.	Runs Used	Run Time (hr)	Atomizing Gas	Feed Type	Comments
1	DSC-4	19	Air	PW-2	Fairly extensive internal erosion on one side.
2	DSC-5	8	Steam	PW-2	Extensive internal erosion with enlargement of the orifice.
4	DSC-2	20	Air	PW-1	Presently in hot cell; as far as can be observed the orifice has not been enlarged.
	DSC-3	29	Air	PW-1	
	SS-2	60	Air	PW-1	
5	DSC-5	24	Air	PW-2	Some internal erosion, fairly uniform.

Spray Solidification Demonstration

Failure of the melter furnace during startup postponed run SS-3. The resistance heated Kanthal A-1 furnace had only 500 total hours of operation at the time of failure. Subsequent visual inspection of the furnace has shown no cause for the failure. The

melter was removed and appeared in perfect condition. The inside of the melter furnace was clean. Overheating or malfunctioning was not evident at any point which could be inspected.

During the startup checkout of the spray calciner it was necessary to flush the atomizing air line to the spray nozzle with nitric acid to clear an obstruction. After flushing, operation appeared normal, but the furnace failure prevented startup of the run.

Product Evaluation (WSEP)

Measurements on three calciner pots (PC-1, PC-2, PC-3) and two spray solidification pots (SS-1 and SS-2) showed no measurable change in the original diameter of the pots which are now filled with radioactive solids. The product evaluation program calls for measuring the pots after they are filled, to see if any pot deformation has occurred. The diameters of pots from runs PC-1, PC-2, PC-3, SS-1, and SS-2 were measured in cell by using large, manipulator-held outside calipers. No deformation was observed in any of the pots; however, it is estimated that the measurement technique used would not detect changes in diameter of less than about 3/16 inch. A satisfactory in-cell measurement technique for measuring pot length has not yet been developed.

Heat transfer calculations for WSEP pots have normally not included any provision for the effect of gamma energy loss, that is, the gamma energy which is converted to heat outside of the pot. A short study was made to see whether significant error would be introduced into the calculations by ignoring gamma loss. It was concluded that about a 2% error might result; an insignificant error in view of the accuracy with which other values pertinent to the calculations are known.

Laboratory Scale Glass Equipment

Equipment for the laboratory scale production of glasses (on a continuous basis) from calcined waste solutions has been assembled. Preliminary runs showed the feed mechanism to be inadequate. A vibratory type feeder will be evaluated. This equipment will be used for evaluation of long-term effects of melter conditions on platinum and platinum alloys.

Solids Storage Engineering Testing

The second phase of design verification tests on environmental test pod #1, demonstration of remote mechanical assembly, has been completed insofar as required to define assembly problems with the pod. The primary results are:

1. Ceramic thermocouple connectors will be used for heater rod connectors for future pods instead of Transite-supported connector lugs.
2. Additional thermal shielding and cooling will be provided for the pot thermocouple connector.
3. Various details on the pot support flange were changed to facilitate remote assembly.

It was concluded from first and second phase tests that, with minor modifications, the first test pod is ready for in-cell service.

Developments on the Solids Storage Engineering Test Facility include:

1. Revision #1 to the design criteria has been submitted and approved. This revision specifies (1) addition of four upper-level lights, two upper-level manipulator sleeves, and process sewer and cubicle interconnections; and (2) modifications to A-cell doors for pass-throughs and for changes in the door opening sequence. The impact of these revisions on the design schedule has not been determined.
2. Bids on the shielding window have been reviewed. Apparently none of the vendors can meet the required shipping date. Negotiations are under way to revise the requirements so that the frame shipping schedule (Sept. 15) can be kept, but the window shipping schedule (Oct. 15) relaxed.
3. The feasibility of in-the-wall coring to extend piping services to the third floor cubicles has been verified. In-the-wall piping eliminates both in-cell piping and shielded gallery piping.

It is expected that actual project work will begin by July 15, with in-the-wall coring followed by installation of piping and, possibly, start of the wall liner.

Intermediate Level Waste Treatment

Efforts on resolving the question of disposing of the aqueous sulfate waste produced in the phosphate glass process have been devoted this month to data analysis. Data on fission product DF between the melter and condensate is meager and varies over a broad range. It has been concluded that a range of ruthenium DFs from 10 to 100 must be considered and that a range of DFs for the other fission products of 10^3 to 10^4 must be considered.

Integrated dose rates for infinite-time exposure for incorporation of the sulfate waste in asphalt were calculated. Without

additional decontamination, the asphalt volume required, if integrated dose is limited to 10^8 rads, would amount to 233 to 2330 gal/MT of fuel for the Ru and 27 to 274 gal/MT of fuel for the other FP's. Obviously, a substantial further DF will be required for any process that proposes final disposition of the waste in asphalt if a reasonable volume is to be realized.

The minimum volume of asphalt as limited by solids content for just the sulfate content will be on the order of 14 gal/MT. Comparing this with approximately 10 gal of glass product/MT indicates the magnitude of the problem.

Fission Product Aerosol Containment

Removal of Organic Iodides with Hydrazine (L. C. Schwendiman)

Removal of methyl iodide from a steam containing atmosphere by transport to a wall wetted with hydrazine was further explored. An aqueous scrub solution of 5% hydrazine - 5% ammonium hydroxide was directed down the vertical walls of the 3.6 m^3 spray chamber which contained a steam atmosphere to which had been added 0.1 mg/m^3 of traced CH_3I . The flow rate of 2.5 gpm and hydrazine concentration were about equal to those used in previous spray experiments. Results of this experiment showed a rate of removal of methyl iodide about the same as for the spray run using the same volume throughput of hydrazine. This result would have been predicted from the previous experiment in which the removal of CH_3I by the wall film was about as effective per unit volume of hydrazine as was the removal from the sprayed hydrazine. Removal half-time of 57 minutes in this experiment was about the same as found earlier for the previous spray run. The methyl iodide depletion curve indicated that removal occurred at two rates. The initial portion of the curve follows a slope consistent with an assumption of a partition coefficient of five which is undoubtedly too high for a pure water system alone at these elevated temperatures. The slope of the lower portion of the curve is consistent with a partition coefficient of two to three which is more in line with recent estimates of the distribution of methyl iodide between water and air. The amount of methyl iodide removed was 95% in the 6-hour run.

The rather minor dependence on factors other than liquid throughput for a given hydrazine concentration gives further evidence that the reaction rate in the liquid phase may be limiting the removal rate under the conditions examined to date. Very fine sprays and higher concentrations of hydrazine will be used in forthcoming experiments.

Higher temperatures and pressures than those attainable in the 3.6 m^3 spray chamber are necessary to simulate the conditions following a reactor accident. The applicability of the tank used in

the CSE simulant development tests (ADF) was studied for these experiments. The durability of the phenoline paint under the atmospheric conditions to be imposed was briefly studied using painted steel washers. At the higher temperatures and pressures, both with water and with dilute hydrazine solution, the paint peeled away from the sharp edge of the washer. Further testing is planned. Other problems relating to the modification of the ADF chamber for spray runs were considered.

Physical Chemistry of the Hydrazine-Methyl Iodide Reaction

(L. L. Burger)

The partition coefficient of methyl iodide between the gas phase and an aqueous solution was determined between 4.85 and 68.5 °C. A gas chromatographic technique was employed to determine the gas phase concentration of methyl iodide which varied from 0.64×10^{-8} to 2.8×10^{-7} g mole/liter. The aqueous concentration of methyl iodide was estimated by a material balance of the amount of methyl iodide introduced to the system.

The partition coefficient $\frac{(\text{g moles CH}_3\text{I/liter, liquid})}{(\text{g moles CH}_3\text{I liter, vapor})}$ was found to be approximated by the relationship

$$\log(\text{partition coefficient}) = -4.82 + \frac{1597}{T(\text{°K})}.$$

The data deviate from this linear relationship about 30 °C, i.e., a positive deviation at 45 °C of ca. 20% and a negative deviation at temperatures above 60 °C. A value of 0.2 for the partition coefficient at 90 °C is given in ORNL-4071 which is only 36% of the value predicted by the equation.

No effect of concentration was found from 6.4×10^{-9} g moles/liter to 1.28×10^{-7} g moles/liter at 29.9 °C. However, the value of 2.75 ± 0.20 for the partition coefficient which is found in this work differs significantly from the value of 3.4 which is calculated from the data reported by Glew (Discussions Faraday Soc. 1953, 150) for solutions with ca. 10^{-2} mole CH₃I/liter.

Geophysical Exploration of Rattlesnake No. 1 Well (W. A. Haney)

Field investigations of the geophysical and hydrologic characteristics of the Standard Oil Co. #1 Rattlesnake Hills Test Well were completed on June 10, 1967. The well was plugged to pre-test conditions on June 12th, and the site abandoned.

Eight different wire-line logging tools were used to log the well and side-hole cores were taken at various locations in both the basalt flows and interflow zones. In addition, seven drill-stem tests were made to evaluate formation transmissibilities and fluid potential.

A meeting was held with Schlumberger Well Surveying Corporation personnel in Houston, Texas, to discuss in detail the interpretation of the electric log data. Five specific zones of interest were analyzed in detail by Schlumberger to illustrate the techniques available and the type of information that can be obtained. Quantitative information can be derived as follows: formation density and porosity, degree of fracture, seismic velocity characteristics, and resistivity characteristics. Qualitative information can be derived as follows: existence of formation fluid as opposed to invasion of drilling fluid, presence of formation gas and indication of permeable zones. Drillstem test data are presently being analyzed as are gas and water samples obtained during the testing.

Side-wall cores taken from 60 separate levels have been given a preliminary examination. Twenty-nine, well-formed cores and 22 samples of mud or partial recovery were obtained. Nine sample attempts were unsuccessful with no core recovery achieved. All of the side-wall cores that exhibited a high degree of recovery were taken from interflow zones. Initial examination of the samples indicates that these interflow zones are mostly weathered and altered basalt rather than volcanic tuff as had been assumed previously.

Three thin sections from the drill core taken by Standard Oil Company at 8084 to 8100 feet were sent to Dr. Glen Bennett of the Washington State Division of Mines and Geology for examination. His analysis states that the core is from a basic augite andesite similar in appearance to the Fife's Peak Andesite which outcrops approximately 25 miles west of Yakima. He also states that an alteration effect is visible that indicates a contact metamorphism by solutions released from a plutonic intrusion into andesite. This information is quite significant because it could indicate that: (1) the Oligocene-Miocene volcanics of the south Cascades extend considerably farther to the east than previously suspected, (2) the source for all the igneous flows underlying the Hanford Project may not be to the south and southeast, and (3) the structural origin of the Columbia Basin anticlines may be related to intrusive activity.

Disposal of Reactor Off-Gas Into Soil Systems (W. A. Haney)

Computer calculations were completed for obtaining minimum travel times as functions of well depth, perforated length of injection structure and injection pressure. A meeting is scheduled at NRTS early in July to resolve data collection requirements and accuracies for analysis of pre-test injections.

Columbia River Sedimentation Studies (D. R. Kalkwarf)

Samples of bottom sediment, collected in the Columbia River estuary, were found to contain Zn-65, Co-60, Sc-46, Mn-54, and Cr-51 at concentrations which varied greatly between points of collection. After fractionating the samples with respect to particle size, it was found that the finer particle fractions contained the greatest concentrations of radionuclides. Interstitial water from tidal flats in the estuary was also analyzed and found to contain extremely low concentrations of radionuclides. Analysis revealed that they were derived in greater part from fallout rather than from reactor effluent.

Simulation Modeling of Expected Thermal Generation
in Selected River Systems (R. T. Jaske)

Field investigations were made in order to verify any physical assumptions made in studies on both the Illinois River and the Deerfield River in Massachusetts. Persons cooperating in the studies were contacted, and photographic records made of pertinent installations and features of both systems.

Two thermographs which were installed at the request of this program, one each at Dresden Island lock and dam and at the Starved Rock and dam, were calibrated using precision apparatus. Data from these installations are now coming in; these also include the daily river flows. In addition, arrangements have been concluded with the Weather Bureau for current data from the Chicago-Peoria, Illinois area. These data are being assembled in order to permit a convincing real time test of the digital simulation system for application to the balance of the study areas under consideration.

Extension of the study on the Illinois River down to mile 80, 190 miles below the Dresden station is currently under way. The lower reach of the river involves additional heat inputs from fossil fuel stations at several points along the reach. Since the simulation program is designed to handle multiple heat inputs, no problems are expected from this portion of the study. Peoria weather data are to be used in these runs.

The Weather Bureau has indicated shipment of the Albany weather record after a 45-day delay. Receipt of these punched cards will permit continuation of the verification runs on the Deerfield River. All other necessary information is on hand. The New England Power Company has indicated a great deal of interest in the results of the computations.

Pressure Vessel Crack Monitoring (J. C. Spanner)

Detection of Metal Overstress by Acoustic Emission

The schedule 189-A form supporting continuation of the program for application of acoustic emission to monitor flaw movement in reactor pressure piping was revised and reissued to include recent changes in program emphasis.

Comparison of data from the latest tests using small pressure vessels, formed from 3" diameter pipe, with that from earlier similar tests points to the surface layer as being very significant in announcing deformation. The earlier tests using "as-received" pipe with the oxide layer in place produced a strong continuous signal as well as transients during the deformation phase of the test. The latest tests used a similar specimen with the exception that the outer surface was sand-blasted to remove the oxide layer. Data from these tests showed the transient signals quite clearly, but there was little evidence of a continuous signal attributable to deformation. It appears that as a deformation dislocation breaks through the surface, the brittle oxide layer acts as an amplifier by adding its own emission to the original signal from the dislocation movement. Further testing is in progress to confirm the indicated surface layer effect. These tests will also endeavor to clarify the predominate acoustic emission transmission mode, i.e., surface waves or shear waves--by varying transducer mounting position.

Data from the latest tests showed frequency components at about 250 kHz. Upon further study, it appeared that this was a resonance excited in the pipe wall. This is one more piece of evidence in support of the earlier conclusion that many of the original acoustic emission signals are in the form of a very short period transient.

Analysis of acoustic emission information using two types of digital analyzers at the Naval Ordnance Laboratory in Corona, California, produced a transient signal analysis quite similar to that derived earlier from selected data using an analog analyzer. One of the digital systems operates in real time and is capable of providing continuous spectral analysis of signals between 10 kHz and 100 kHz, with the output recorded on 35 mm film by intensity modulation. There are other types of output better suited to the study of acoustic emission; however, this output is quite adequate for evaluating the analysis technique. Each of the burst tests were analyzed with this system. The other spectrum analyzer was not capable of real time analysis but did provide useful information about signals in selected areas of the burst tests. The frequency response of this system is 1 kHz to 10 kHz with this spectral information recorded on 4 x 5 poloroid film. The analysis frequency range was extended by slowing the tape playback speed by a factor of 16. Data from a burst test were inspected with this slower

system. Although the results from digital analysis correlated closely with earlier analog analysis, both digital approaches to spectral analysis offer important advantages over the analog method employing tracking filters. The most important feature is their ability to analyze random-transient signals. The tracking filter requires a much higher signal repetition rate in a given period of time to produce a measurable indication which makes it poorly suited for analysis of transient data.

The emission rate analyzer was used for preliminary study of acoustic emission generated by the metal fatigue process with encouraging results. Two types of fatigue were monitored; a zirconium-niobium tube failed by cycling the internal pressure (stress) in the positive direction only, and carbon steel cantilever beam specimens failed by reverse cycling the stress in bending. Both types of test showed similar results, i.e., the emission rate increased early in the process, dropped off during the middle part, and then showed a steady, pronounced increase starting well before failure and continuing until failure occurred.

Preliminary work is in progress to evaluate a new data interpretation technique which will produce both count rate and frequency information. This approach will be relatively inexpensive and straightforward if it proves to be functionally satisfactory.

Two more multilayer electrostatic transducers suitable for evaluation have been produced locally. These are now being lab tested prior to using them for monitoring acoustic emission test data. Alignment of electrode deposition masks has presented a significant problem in fabricating these transducers. Photo-resistive etching techniques are being investigated as a possible method for obtaining better uniformity in the masks. Three electrostatic transducers being fabricated by Thermionics, Hayward, California, are still in process.

Work is in progress to produce a prototypic high temperature piezo-electric transducer for evaluation. The first model will use a PZT-5 crystal for expediency to permit testing the other transducer components at 600 °F, the intent is to ultimately incorporate a higher temperature piezo-electric material such as lithium niobate.