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AEC
RESEARCH
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DEVELOPMENT
REPORT

**PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
FOR JANUARY 1967**

**DIVISION
OF
REACTOR DEVELOPMENT AND TECHNOLOGY
PROGRAMS**

FEBRUARY, 1967

BATTELLE-NORTHWEST

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Nuclear Technology
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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

FOR JANUARY 1967

AEC DIVISION OF
REACTOR DEVELOPMENT AND TECHNOLOGY PROGRAMS

By

The Staff of Battelle-Northwest

S. L. Fawcett, Director

February 1967

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

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SUMMARY

CIVILIAN POWER REACTORS

Nuclear Systems and Concepts Analysis

Calculation of a nuclear generating system containing five reactor types has been completed in preparation for a system containing 30 or more reactor types being decided by the System Analysis Task Force.

Fuel burnup histories from each reactor type are being distributed by the System Analysis Task Force to each of the reactor task force involved.

Conceptual Reactor Design Studies

All but four volumes (on Southwest streams) of the U. S. Geodetic Survey Water Supply Papers have arrived. Flow and temperature correlations are continuing for Pacific Northwest streams.

R. W. Beck and Associates have essentially completed their studies on heat dissipation system costs, and results are expected to be received the week of January 30.

The three portions of the design analysis of the steam-cooled fast reactor: heat transfer, pressure drop, and cycle efficiency have been successfully combined into the design simulation of the reactor. Additions are now being made to the basic simulation.

The fuel cycle cost portion of the economics simulation is about half completed.

HTGR Graphite

The apparatus has been assembled to measure the surface area of small graphite samples by drypton adsorption at liquid-nitrogen temperatures.

Computer calculations of the rate of graphite oxidation and steady-state gas compositions are being initiated for a typical high-temperature gas-cooled reactor.

USAEC-AECL Cooperative Program

Both control and in-reactor creep tests are in progress to determine creep rates in the 300 to 400 C temperature range for stresses from 36,500 to 55,000 psi.

A defect in the analog simulation of the BLW-250 has been found and corrected. Further checks of the system are being made before rerun of the earlier cases.

A report describing the analytical phase of this program is being prepared for issue. This report includes detailed descriptions of the mathematical analysis of cross-flow mixing and the computer program (COBRA) used in the solution of the equations describing the mixing process. The final assembly was started on the test section to measure the turbulent component of cross-flow mixing during boiling. The results of these tests will be combined with earlier tests on boiling diversion of the coolant flow to provide input data for the computer program COBRA and to serve to verify the analysis.

Microhardness data have been obtained from coupons representing three different heat treatments and irradiation levels in the ETR.

Postcleaning weights are reported for G-7 loop specimens exposed four cycles in the ETR G-7 loop. Crud deposition on 2-cycle and 4-cycle G-7 loop specimens is summarized.

APPLIED AND REACTOR PHYSICS

Plutonium Criticality Studies

A new series of criticality experiments were begun with PuO₂-polystyrene compacts and the remote split-table machine. The experiments are a continuation of the basic research program to provide data needed for evaluating the effects of moderation and Pu-240 in intermediate neutron spectrum plutonium systems. The fuel being used in the current experiments has an H/Pu atomic ratio of 5 and a Pu-240 isotopic content of about 12 wt%. A critical mass of about 30 kg Pu was obtained for a Lucite reflected assembly of near cubic geometry.

Phoenix Fuel Reactor Program

One recording of reactor noise from the CAF-Phoenix experiments has been successfully analyzed. CAF-Phoenix calculations are now within about 1.5% in k of the experimental value.

The PRCF-Phoenix core has been installed in the reactor cell, and all fuel assemblies are ready for use.

Expressions of interest to fabricate the Phoenix fuel load for the MTR experiment have been received from commercial sources.

The use of borated end fittings on the MTR-Phoenix fuel elements to suppress the power peaks shows promise. An improved control rod model indicates a shutdown k of between 0.81 and 0.86 for the Phoenix fuel loading in the MTR (without borated pieces for power peak suppression).

The calculated results for Phoenix fuel in the HFIR continue to make this application look interesting.

A preliminary examination of Phoenix fuel in the PRTR indicates a region of possible fuel loadings of interest.

High Temperature Reactor Physics Studies

The Acceptance Test Procedure on the reactor heater control system was completed. The reactor shell is being closed for pressure and leak tests.

The gas blower has been tested and accepted after minor adjustments.

The neutron generator was run for four hours and accepted. A vertical flux traverse was made with gold foils.

The control rods were assembled. The rod position feedback problem has disappeared.

Work on Criticality Safety Specifications, Operating Procedures, Design Tests, and Operator Qualification Tests continued.

The temperature coefficient of the 7-1/2-inch graphite lattice to be used at startup was calculated using two current calculational procedures and compared with a previous exponential experiment.

The assembly of the driver fuel and flux adjusting fuel was completed.

Questions from RDT on the Safety Analysis Report have been answered by letter, to be sent through the Richland Operations Office. Some additional drawings and revised pages for the SAR are being prepared.

REACTOR FUELS AND MATERIALS

Fast Fuels Oxides and Nitrides

Die materials have been selected, a press is on hand, and equipment is being designed for use in vacuum hot pressing of high density UN-PuN pellets for irradiation testing.

The facility for producing small batches of mixed nitrides is in operation. Preliminary runs indicate that the facility is capable of producing about 250 grams of mixed nitride pellets per week on a one shift/day basis.

Basic Swelling Studies

Capsule 36 (450 C, ~30 psi, ~0.35 at.% BU) was discharged after a successful irradiation. Capsule P-15 (700 C, 1000 psi, 0.2 at.% BU) was charged and is operating successfully. Construction on two additional capsules (P-15 and P-16) is almost complete.

The electron microscope examination of uranium specimens irradiated to 0.2 at.% BU at 610 C (alpha phase) and 690 C (beta phase) showed no change in structure as the result of annealing the specimens at 625 C for 100 hours.

The modification of facilities and the development of techniques and equipment to postirradiation anneal uranium at temperatures to 900 C and pressures to 15,000 psi are partially complete.

Nondestructive Testing

An alternate design for nil-ductility-transition irradiation specimens is being evaluated in order to maintain specimen temperatures below a maximum of 550-600 F. Preliminary studies indicate that it may be possible to obtain two different irradiation temperatures from one capsule loading through judicious placement and selection of helium gaps.

Additional data were obtained for use in a study relating single frequency, multifrequency, and pulsed eddy current nondestructive testing techniques. Test coil output signals were measured for several different test conditions which showed the relative effects of varying test coil diameter and test frequency. Lower test frequencies were found necessary in order to reduce the parasitic loading effect of the unused test coils in the probe.

Recent performance tests on the sinusoidal thermal wave instrument have shown that long term stability is not yet within acceptable limits. A study to further investigate and correct the causes for these instabilities is in

progress. Development is continuing on the remote transient technique for measuring surface thermal impedances using a high intensity xenon flash lamp.

Nuclear Ceramics

Single crystal specimens of UO_2 , ThO_2 , and polycrystalline UO_2 , ThO_2 , and UO_2 - ThO_2 specimens were prepared, characterized, and distributed to domestic and foreign laboratories for use in basic studies and information exchanges.

The thermal diffusivity of $(\text{U}_{0.8}, \text{Pu}_{0.2})\text{O}_2$ was measured from 100 to 1500 C using a laser-pulse thermal diffusivity apparatus.

A ternary evaporator for depositing compositions corresponding to isothermal sections of the Pu-O-N pseudo-ternary phase diagram onto platinum substrates was assembled and is being calibrated.

The electrical conductivity of UO_2 above 1400 K can be represented by the equation

$$\sigma = 2.104 \times 10^{-2} T^{1.401} e^{\frac{-0.916 \text{ eV}}{kT}} \text{ mhl/cm}$$

Two mixed uranium-plutonium oxides were examined by optical and electron microscopy.

Nuclear Graphite

The high-temperature graphite capsule, GEH-13-13, is operating satisfactorily, but the maximum temperature of 1200 C is 150 C below the design value. A local flux depression is causing the lower temperature.

Irradiations of graphite pins to 3×10^{22} and 6×10^{22} nvt ($E > 0.18 \text{ MeV}$) at 550 C have begun in EBR-II. Tentative approval was secured for a seven-pin subassembly irradiation in Row 7. This joint irradiation with ORNL will permit the attainment of higher irradiation temperatures.

Samples from the GETR irradiation capsule, H-3-13, provide data to 2.4×10^{22} nvt ($E > 0.18 \text{ MeV}$). The graphite, NC-8, at 800 C displays net length changes of +19.8% and -7.5% in the transverse and parallel directions, respectively. At 800 C changes in CSF were +10.1% transverse and -7.8% parallel. Both materials now seem to be expanding in the parallel directions.

The irradiation of "dimensionally stable" graphites in Capsule H-3-24 continues satisfactorily in the GETR. Scheduled discharge is late January.

All irradiations of pyrolytic and single crystal graphite from Union Carbide Corporation have been completed.

A modified constant-stress model complements the modified constant-strain model proposed by Buessem to explain properties of polycrystalline, anisotropic, ceramic materials. The two models closely bracket experimental values of thermal expansion coefficient and Young's moduli of some graphites. This good agreement implies that the models should be useful in predicting or explaining other properties.

Irradiation Damage to Reactor Metals

Fabrication of a program heat of Incoloy 800 is essentially complete. Plate, bar, sheet, and tube rounds will be shipped after final inspection of the tube blanks.

A liquid-sodium, irradiation capsule will soon complete 90 full-power days in the ETR for an estimated 1.3×10^{21} fast fluence.

A complete creep-rupture curve for Hastelloy X-280 was obtained from an in-reactor test conducted at 1000 C temperature and 3500 psi stress. Specimen rupture occurred in much less time than predicted from out-of-reactor data.

Electron microscopy examinations of stainless steel creep samples are still in progress, and additional studies are being initiated on tensile specimens irradiated and tested at various temperatures. Capsules have been designed for conducting irradiations in the G-7 loop at temperatures greater than 290 C.

Stress-rupture tests on eight irradiated Inconel 600 specimens are nearing completion, and similar tests on seven irradiated Inconel X-750 specimens have been initiated.

A program to determine the effects of irradiation on Sintered Aluminum Products (SAP) in support of the HWOOCR program is being initiated. Sample quantities of XAP 005 and SAP 895 materials have been received from Atomics International (AI). Metallographic examination of this material is now in progress. Two sheath capsules designed for irradiation in the G-7 hot water loop facility are being manufactured. These capsules will be placed in the G-7 loop with thermal monitors to evaluate the heat transfer characteristics of the design.

Specimens of titanium, niobium, niobium-1% zirconium, and Zircaloy-2 were exposed 37 days in the ETR G-7 loop in pH-10 NH_4OH , <0.05 ppm O_2 at 270-280 C. Titanium weight gains were nearly a factor of 10 lower than Zircaloy-2 weight gains. Weight gains were high and variable on niobium and niobium-1% zirconium. However, the niobium-bearing specimens did not absorb hydrogen in-flux, while titanium did absorb 0.69 mg/dm^2 .

The monitor materials for the counting laboratory intercalibration test have been irradiated and are being counted. These materials will be distributed to participating laboratories early in February 1967.

Fracture toughness measurements on A302-B pressure vessel steel irradiated to 4×10^{19} nvt ($E > 1$ MeV) at 280 C in water reveal a significant reduction in toughness below the ductile-to-brittle transition temperature. Although the material suffered embrittlement due to irradiation, the extent of crack propagation was less because the difference between the fracture toughness and the stress intensity at crack arrest was small.

Experiments to gain insight concerning the influence of side grooves in crack-line loaded specimens have revealed that the measured fracture toughness can be abnormally large if the side grooves are either too deep or too shallow. This effect is in agreement with calculated variations in stress concentration as a function of groove depth.

Two EBR-II subassemblies containing FFTF candidate alloys, AISI 304 and 316 stainless steels, together with backup alloys, have been completed. These subassemblies consist of sodium-filled subcapsules designed to operate at 900, 1200, and 1400 F (480, 650, and 750 C) during irradiation. Target fluences are 1×10^{22} and 1×10^{23} nvt.

Tensile tests on specimens of AISI 304 stainless steel irradiated to 2×10^{22} nvt in the EBR-II are in progress.

The first phase of Building 321-A renovation is nearing completion. Construction of a shielded prototype cell has proceeded far enough for a radiation leak test. A review of all requirements indicates it may be possible to operate this cell with radioactive specimens before building renovation is complete. An information package is being prepared for offsite procurement of the piping to service the liquid metal loop creep rigs. The liquid metal loop is now under construction with shipment expected in mid-February. The creep test units for use in the loop are in final design. A device for accurately measuring and dispensing liquid sodium to the various test units has been designed and is being built.

A series of liquid-metal and inert-gas, ex-reactor aging studies is being prepared. These experiments will duplicate environmental histories of several irradiation experiments in progress.

Techniques have been developed that will permit the use of nonequilibrium permeation techniques for surface effect studies of hydrogen behavior in cladding materials.

The model gas loop has operated a total of 2197 hours since installation of the new heater; this includes 117 hours during the past month. The loop was shut down most of the month while test specimens were being examined. During the shutdown the vertical test specimen assembly was reinstalled to determine pressure drop across this test assembly at various flows and temperatures.

The scoping of an auxiliary heater to increase specimen temperature in the loop has been completed. Detailed design of this heater has been started.

Temperatures of a columbium hanger rod and test section liner were calculated for a set of "worst case" conditions in the ATR helium loop. The hanger rod strength at a calculated peak operating temperature of 2530 F (1390 C) will be investigated. Additional hanger rod and liner temperatures will be calculated as a function of helium temperatures, materials, and heating rates.

Experimental pressure drop measurements in the model gas loop indicate that the maximum allowable pressure drop for the ATR specimen test assembly will not be exceeded with the current design.

Corrosion coupons exposed at temperature for 504 hours in the ATR model loop continue to show the weight change trend previously reported. Micrographs of Hastelloy X-280 and Haynes 25 illustrate that evaporation accounts for their weight loss. Niobium shows an impurity case accounting in part for its weight gain.

A topical report describing efforts to purify Mo has been completed. The first experiment to study dislocation multiplication during yielding of metal single crystals by ultrasonic attenuation has been performed on a single crystal of Mo. The equipment appeared to be operating satisfactorily and showed a small increase in attenuation in the elastic region and a rapid increase in the pre-yield, microstrain region. The test was terminated when the epoxy bonding of the crystal broke. Refinements in the mounting technique must be made before tests in the yield region can be completed.

ATR Gas Loop Operation and Maintenance

Vendor data on the ATR gas-cooled loop are being reviewed and comments forwarded to the architect-engineer.

Inspections methods are being developed on the analytical instrumentation to assist in proving that the vendor is able to meet the strict requirements for gas analysis. PNL will direct and review the final acceptance of these instruments.

Continual surveillance on loop components by PNL has resulted in better quality control. This effort has uncovered problems in welding that do not meet the requirements of the Nuclear Code.

Metallic Fuels Development

Irradiation of three tubular fuel elements of Th - 2.5 wt% U - 1.0 wt% Zr clad with Zr-2 continued in the P-7 hot water loop at the ETR. The maximum exposure element has achieved an exposure of 2 at.% BU (~17,000 Mwd/ton). Three other irradiated elements are being stored in the ETR basin.

The irradiation of 24 Zircaloy-2 clad hollow core uranium fuel elements 0.45-inch diameter by 6.25-inch long continued in the ETR M-3 loop facility during cycle 86. The maximum exposure at the end of cycle 86 is approximately 2000 MWd/ton. Plans are being made to perform nondestructive neutron radiography on selected elements.

Fast Reactor Dosimetry and Damage Analyses

Fast reactor dosimetry experiments were conducted in the EBR-II during a low power run. The data are being analyzed.

A study is being conducted to develop high temperature ceramic fluence monitors suitable at temperatures above 1200 C. The first of these monitors has been fabricated and is being tested in-reactor.

ENGINEERING DEVELOPMENT

Neutron Flux Monitors

Test program authorizations and capsule design information are being obtained for planned ETR in-core testing of regenerative thermal neutron flux detectors.

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

A completed microwave assembly, including the input waveguide and resonant cavity, was successfully tested during initial experiments regarding measurement of high in-core temperatures.

Upstream Boiling Burnout

A new series of tests to study upstream boiling burnout and related phenomena was started. In tests performed with coolant velocity of 3×10^6 and a system pressure of 1500 psig, a flow pattern change was indicated by thermocouples installed along the test section. This pattern change occurred first at the downstream end of the test section at approximately 20% below boiling burnout heat fluxes. It was subsequently observed to move upstream on further power increases. This flow pattern change is believed to be a possible cause of the upstream boiling burnout phenomenon.

PLUTONIUM UTILIZATION PROGRAM

Fuels Development

Underwater equipment items for the examination of FERTF fuels have been procured and/or fabricated.

Results from recent critical tests indicate that the best power match between the FERTF test element and the Batch Core elements in ring 1 can be obtained using an 8-rod cluster design for the FERTF test element.

Fifty-five fuel elements in PRTR were examined before Batch Core power tests and found to contain foreign material lodged between fuel rods. To eliminate potential problems which could result from such material, all elements (70) that were in the reactor during the outage were returned to the Plutonium Fuels Laboratory for disassembly, cleaning, and rebanding prior to recharging.

A simpler and more reliable fission gas pressure sensing system was designed and is being fabricated.

Gamma and x-ray energy spectra analyses were made on plutonium fuel from the Yankee reactor. The americium buildup even after short periods of time appears to be the main source of radiation.

One 50-gram and three one-kilogram plutonium sources were received for neutron dosimetry measurements from various fuel sources producing greater than 20% Pu-240. Reject Phoenix Pu-Al fuel plates were received for neutron dosimetry measurements.

Cost studies have been completed for PuO₂-UO₂ manufacturing plants with capacities of 1/4- and 1/2-ton/day of fuel.

Reactor Physics

Calculations of photoneutron production in D₂O moderated reactor systems have been completed.

Analytical correlations of burnup data obtained from Pu-Al fuel irradiated in the PRTR have been made.

Experimental and computational studies have been conducted to improve the power match between a FERTF element in channel 1550 and HPD elements in the first ring of channels in PRTR.

Improvements are being made to the PNL Gamma Scan Facility in preparation for studying fuels irradiated in a fast reactor environment.

The ratio of Pu-241 decay to Pu-241 loss by neutron absorption during the irradiation of Pu-Al fuel in the PRTR has been determined.

Analytical correlations of UO_2 - 2 wt% PuO_2 fuel in H_2O moderator experiments have been made. The effects of moderator voiding and moderator height on k_{eff} have been studied.

Analytical correlations of the Saxton fueled experiments conducted at the Westinghouse Reactor Evaluation Center have continued. The studies included single zone and two-zone loadings of PuO_2 - UO_2 and UO_2 fuels.

Four fuel rods containing 350 micron PuO_2 particles in UO_2 pellets and five rods containing PuO_2 - UO_2 solid pellets were completed.

Measurements of the effect that the PuO_2 particle size has on reactivity were started in the PCTR. Results of the initial experiment show a decrease in reactivity for an increase in particle size from 0 to 350 microns.

Recent calculations indicate that 2 wt% mixed oxide experiments over a fairly wide range of lattice pitches can be performed in the PCTR with the proposed water tank assembly.

Two man-years of theoretical physics work on generalization of least-square fitting logic, to meet the needs of the Plutonium Recycle Program, progressed to successful completion during the past month.

Two Battelle representatives from Theoretical Physics participated in a three-day conference on Synthesis Methods for Burnup Analysis, sponsored by the Argonne National Laboratory.

A variety of codes required for the processing of nuclear data were converted for use on the Univac 1107 computer.

A code to produce HRG data tapes in decimal card image for general exporting was computed.

Numerical techniques associated with calculating thermal neutron scattering cross sections from theoretical models were improved by the development of a new computer program named DELERF.

The evaluation phase of the RBU code development program has been completed.

Reactor Engineering Development

Recently, a rupture disc had broken out of the primary system block valve, and particles of this disc had spread throughout the primary system. An analysis was performed to determine the possibility of fuel overheating resulting from these particles lodging in the inlet piping or in the fuel elements. It was concluded that particles lodged in the fuel elements would not cause fuel temperatures to increase sufficiently to result in melting of the cladding but could be high enough to cause a cladding rupture.

Materials Development

The 300 C burst stress of a specimen from discharged PRTR pressure tube 5696 was 81,000 psi at a calculated fluence of 2×10^{21} , $E > 1$ MeV. This stress represents a continuing and expected increase in strength with exposure. Another point obtained on a plot of failure strength of a pressure tube containing a 3/4-inch long axial flaw vs test temperature reveals a -10^4 psi/ $^{\circ}\text{C}$ linear decrease in strength with temperature up to at least 300 C.

Kinetic studies of the OPG dissolution of sintered UO_2 show that the dissolution mechanism is different from that found in HNO_3 . Reaction products catalyze the HNO_3 dissolution of UO_2 , and, consequently, the greatest dissolution rate occurs under quiescent conditions. In the OPG solution the mechanism is simple dissolution and flow past the UO_2 increases the rate.

The P-4 valve rupture disc was discovered to have failed at least six months previously. Alpha contamination on the disc was analyzed. The primary system was flushed through fine-mesh screens, removing principally ion-exchange resin, metal fragments, and oxide flakes.

Sixty tubular Zircaloy-2 and Zircaloy-4 corrosion specimens were exposed 209 days in the PRTR at a position normally occupied by an outer fuel rod. Coolant conditions were pH-10 LiOH at 246-279 C (475-532 F). Flux-induced oxidation was slight on specimens exposed as-etched. However, specimens pre-filmed to 17-19 mg/dm² at 400 C (750 F) underwent accelerated oxidation, up to a factor of six times the corresponding out-of-reactor rates. Weight gains were relatively high, but acceleration factors were 1.5 or less on specimens prefilmed at 425 C (797 F) to 50-70 mg/dm².

Test Reactor Operation

Approval to operate the PRTR was received on January 11, 1967. Criticality was achieved at 0200 on January 25, 1967, after extensive flushing of the primary coolant system and the usual and special pre-operational and startup checks were completed.

Several critical tests were conducted to evaluate the nuclear relationships of the proposed 6-rod and 8-rod cluster fuel elements for the Fuel Element Rupture Test Facility.

The Batch Core Power Tests were begun on January 25, 1967.

EBWR Demonstration Program

A capsule which operated at a maximum linear power of 12.1-17.4 kw/ft (397-571 W/cm) and to a burnup of 18,200 MWd/ton of fuel (4.52×10^{20} fissions/cm³) released 14% of the fission gas.

Arrangements have been made to use the Yankee single element cask for shipping irradiated EBWR fuel from Argonne National Laboratory to Pacific Northwest Laboratory.

NUCLEAR SAFETY

Containment Systems Experiment

The reactor simulator vessel was installed in its test stand and installation of braces and pipe jumpers started. A three-component aerosol mixture of iodine, cesium, and UO₂ - Zircaloy cladding fume was investigated in a small stainless steel laboratory containment vessel. Gas phase removal half lives were 10 min for iodine and cesium and 15 min for uranium, and no evidence of interaction between cesium and iodine was found. Performance of a Maypack cluster assembly procured for the aerosol experiments in the CSE containment vessel was evaluated in this run, and it was found that undesirably high aerosol deposition in the nose cone resulted from the disturbed flow patterns around the inlet ball check valve. The on-line computer gamma energy analytical system was used successfully for the analysis of aerosol tests, and 170 samples were analyzed in two shifts using only two of the four detector stations. Leakage rate tests continued for the nominal 1%/day containment vessel leakage condition at pressures of 30 and 10 psig.

Pressure Vessel Crack Monitoring

Specimens are being prepared for a preliminary study of acoustic emission effects in internally pressurized vessels. This series of tests is the planned predecessor of a later test to identify the problems of detecting acoustic emission under simulated dynamic field conditions. Three different types of charge amplifiers were evaluated for applicability to the acoustic program, and it was found that charge amplifiers using field effect transistors have better noise level characteristics than bipolar transistor amplifiers.

Masks for multilayer transducer fabrication were completed, and deposition equipment for remote mask manipulation has been received. Work will continue on fabrication and evaluation of multilayer electrostatic transducers because of their potential for further increasing detection sensitivities.

Reactor Safety Analysis and Evaluation

A heavy section steel specimen was prepared, and the arrangements were made for conducting a gamma heating experiment on this specimen in the Sandia Experimental Reactor Facility.

Waste Solidification Condensate Treatment

Laboratory experiments were conducted to evaluate the effectiveness of waste incorporation in asphalt as a method for treating glass melter condensate. Leach tests are continuing.

Fission Product Aerosol Containment

Experimental studies are in progress to determine the drop size and spray patterns for nozzles for the 4 cubic meter aerosol chamber being used for demonstration of the removal of organic iodides with hydrazine. Laboratory studies in parallel with the engineering demonstrations are determining the effects of interfacial area and volume on rates of removal.

Geophysical Exploration of Rattlesnake #1 Well

Many preliminary steps have been completed or are in progress in the efforts to reopen the Rattlesnake #1 Well for geophysical exploration.

Columbia River Sedimentation Studies

Substantial quantities of some radionuclides were dissolved from river bottom sediments during the reactor shutdown period.

Radioactive Waste Solidification

The first radioactive spray solidification run in WSEP was only partially successful. Internal buildup caused by poor atomization forced early termination of the run.

Out of specification waste solution forced testing of sulfate-bearing flowsheets for pot calcination. Sulfate-free tests will be delayed until the next delivery of wastes.

In tests of conditions which may cause poor atomization, poor atomization was obtained using steam as evidenced by cake buildup at the bottom of the calciner.

The leach test procedure developed for leach testing radioactive products from WSEP gave good precision in tests on nonradioactive materials. Leach rate is affected by the thermal history of the product.

Laboratory simulated condensate produced from a phosphate glass process was successfully contained in asphalt. Leach rates are being studied.

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

The Phase II calculations of a five-reactor problem are complete. The results are satisfactory in that they prove that the tools to be used in the remainder of the Phase II and Phase III efforts are well debugged, which was the primary purpose of the calculations. However, the numerical values are not so good because of difficulties in the data, difficulties which have been corrected for later Phase II work. The Phase II calculations of a many-reactor problem are set up and being run at month-end.

Only the plants to be installed from 1970 through 2040 and the energy from them are being considered, resulting in a more concise forecast of additional capacity required for each cost area and time period. A computer code is being developed to accomplish this task more readily should any changes in electrical demand or cost area be desired.

Fuel burnup histories, each covering a 30-year operating period, have been calculated and saved on magnetic tape for 30 reactors. In each case it was assumed the reactor would operate at 85% capacity factor for the first six years of its life and that the capacity factor would decrease 2%/yr thereafter to 37% at the end of useful life. Reactor types included were pressurized water, boiling water, sodium cooled oxide fuel, and sodium cooled carbide fuel. Appropriate selections of enriched uranium fuel, plutonium recycle and plutonium enrichment with exchange between reactor types were included. Amount or level of enrichment was also varied to permit the linear program model of the electrical industry to select the optimum mode of operation of each type reactor.

Reactor Systems Analysis

The output for ORNL's POWERCO is being revised to provide more useful power cost details.

PACTOLUS Calculations

Fuel cycle cost and mass balances have been calculated with the PACTOLUS code for reactor types to be included in Phase II of the nuclear systems analysis effort.

PACTOLUS Calculations

PWR	Uranium
PWR	Plutonium Recycle
BWR	Uranium
HWO CR	Uranium
HWO CR	Thorium metal
HWO CR	Thorium oxide
HTGR	Thorium carbide
HW-BLW	Uranium
FBR-steam cooled	Uranium-plutonium
FBR-helium cooled	Uranium-plutonium
LMFBR	Uranium-plutonium oxide
LMFBR	Uranium-plutonium carbide

Copies of the PACTOLUS calculations are being sent to the individual reactor task forces involved in the analysis of each. Two sets of calculations will be included, one showing the cost for the particular reactor operating in a changing economic environment; that is, changing uranium prices, changing fabrication cost, and changing reprocessing cost throughout the 30-year history of reactor operation. A second set of PACTOLUS results will show the same reactor history analyzed with a fixed economic situation, which is usually the conditions under which the particular reactor design was optimized. Other reactor types of interest which are not included in the above list will be included in Phase III calculations which will constitute the final report.

Conceptual Reactor Design Studies (J. C. Fox)Thermal Sink Limitations Study

All but four volumes of the U.S.G.S. Water Supply Papers on order have arrived; the missing volumes contain data on Southwest streams. Correlations of flow and temperature data for streams in the Pacific Northwest are continuing.

Reactor Siting Study

R. W. Beck and Associates have essentially completed their cost studies on heat dissipation systems. Their work is expected to be completed by the end of January, and results should be received the week of January 30.

Steam-Cooled Fast Reactor

The three portions of the design analysis: heat transfer, pressure drop, and cycle efficiency, have been successfully combined into the design simulation of the reactor.

Clad and fuel temperature constraints are in the process of being included in the simulation. Fission gas plenum sizing and clad thickness determination are also being done at the present time. The effect of surface roughening will be added to the simulation in the near future in such a way that reactors both with and without surface roughening can be included.

A fuel cycle flow diagram, for use in estimating the fuel cycle cost portion of the economic simulation, has been developed. The fuel cycle has been formulated to handle unit costs, such as fabrication costs, in a form that is compatible with the input that will be available. The mathematical relationships describing the fuel cycle are approximately half written. They will be finished and programmed into the simulation early in the next report period.

HTGR Graphite (R. E. Nightingale)

Graphite Surface Area Measurements

The modification of the surface area apparatus to provide the capability for using krypton as adsorbate in the measurement of graphite surface areas has required the calibration of a thermistor gauge for measuring the krypton pressure and the determination of pertinent line volumes. Because the vapor pressure of krypton at liquid nitrogen temperature is only about 2.6 mm Hg, and krypton pressures during a surface area measurements may only range up to a few hundred microns, a special means is needed to measure these low pressures accurately without contributing excessive volume to the system. A thermistor sealed into the vacuum line as one arm in a Wheatstone bridge circuit is a sensitive pressure-measuring device. Variations in the gas pressure and hence the thermal conductivity of the gas are readily detected by the thermistor with its large negative temperature coefficient of resistance. The bridge circuit is balanced at pressures $<10^{-5}$ mm Hg by means of a potentiometer and higher pressures are then measured as an off-balance of the bridge. The response is about 2 mv/micron for pressures less than about 200 microns and decreases gradually with further increase in pressure to a value of about 0.2 mv/micron at 2000 microns. Reasonably accurate pressure measurements can be made up to a pressure of 3 mm of Hg.

Surface area measurements are now being made to assess the accuracy and reproducibility of this technique for measuring the surface area of small graphite samples.

Graphite Oxidation in a High-Temperature Gas-Cooled Reactor

A computer program to simulate graphite oxidation in nuclear reactors is being revised to calculate graphite oxidation and gas concentrations in a typical high-temperature gas-cooled reactor.

The program simulates a system in which the gases flow down a cylindrical channel 0.625-inch in diameter and 271-inch long. The temperatures are varied down the channel to simulate actual reactor temperatures. Conditions will be varied to determine reaction rates and steady state concentrations for channels in the maximum average and minimum power region.

An attempt will be made to assess the effect of various initial concentrations on the rate of oxidation and gas concentrations at steady state. The program has been modified to permit recycling of the gas where the exit gas mixture is returned to the channel entrance after corrections are made for in-leakage of water vapor from the heat exchanger and removal of impurities in the purification system.

The following chemical reactions are considered in the calculation:

The effect of radiation and inert gas sensitization on the latter two reactions is included. Rate constants for the various reactions have been determined experimentally. Possible effects of catalytic impurities on the graphite reactions will be accounted for as an increase in the rate constants. The importance of the catalytic effect will be assessed by choosing a range of rate constants.

Of primary concern is the $\text{H}_2/\text{H}_2\text{O}$ ratio. The effect of initial gas composition, graphite temperatures, and increase in rate due to catalysis on this ratio will be evaluated.

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

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

Both control and in-reactor creep tests are in progress to determine creep rates in the 300 to 400 C temperature range for stresses from 36,500 to 55,000 psi. Specimens which have not been taken to rupture during the in-reactor creep tests are being tensile tested to determine the total elongation. Specimens for which testing is complete are being prepared for metallographic examination.

In-Reactor Measurements of Creep in Zircaloy-2

Capsules are being assembled for in-reactor creep tests on sections of pressure tube material. In one of these tests the specimen will be first pre-stressed at temperature and then subsequently stressed-to-rupture at a higher

stress. Another test will be conducted at the higher stress alone to provide comparative data on creep rate and total plastic strain to failure.

Hydraulic and Nuclear Stability in Parallel Flow Channel Systems

The hydraulics model used in the analog computer study of hydraulic and nuclear stability of boiling water cooled reactor systems was decoupled from the nuclear model and run independently on the analog computer. It was found that a saturating amplifier was disguising the instability threshold. Re-scaling removed the problem and the instability threshold was observed at a power level just over twice the nominal power, consistent with results obtained earlier with the digital computer code, HYDNA. A check of the fuel temperature circuit is currently under way, and a rerun of earlier cases is planned upon final checkout of the system.

Cross-Flow Mixing Between Parallel Flow Channels During Boiling

Both analytical and experimental portions of the program to determine values of cross-channel mixing during two-phase flow in rod bundle fuel elements were continued. The basic purpose of this program is to obtain a method to predict local flow and enthalpy conditions in each subchannel of rod bundle fuel elements. This information will help provide a more accurate means to predict boiling burnout conditions.

A report describing the analytical phase of this program is being prepared for issue. This report includes a detailed description of the development of the mixing equations, the numerical techniques used to solve these equations, a complete listing of the Fortran IV computer program COBRA used in the solution, and a sample problem.

In the experimental portion of this program, final assembly and installation of instrumentation on the new laboratory test section is under way. In the new test section, the flow passages formed by adjacent square pitch array fuel rods are simulated. This test section will be used to measure only the turbulent component of cross-flow mixing during boiling. The flow subchannels have equal flow areas and heat inputs so that two-phase effects during boiling will be the same in each subchannel. Therefore, mixing from flow diversion will be negligible. Measurements of the amount of mixing will be accomplished by injecting a heavy water tracer into one of the subchannels and measuring the tracer concentration in the outlet streams. From the tracer concentration, the amount of mixing of both phases can be determined. Because of the similarity of the thermodynamic characteristics of light and heavy water, it is expected that the presence of the tracer will have negligible effects on the boiling behavior of the test section. A secondary ion (Li) tracer is also going to be used for the experiments. This will check the mixing rates obtained for single phase conditions using the heavy water tracer and also provide a measure of mixing between the liquid phases of the two streams.

Evaluation of Zirconium - 2.5 wt% Nb Pressure Tubing

Microhardness data were obtained from specimens of Zr - 2.5 Nb irradiated as corrosion coupons in the G-7 loop of the ETR. These coupons represent three heat treatments:

- A. Quenched from 850 C, 30% cold worked and aged at 500 C for 24 hrs.
- B. Quenched from 850 C, 20% cold worked and aged at 500 C for 24 hrs.
- C. Annealed at 575 C for 5 hours, and 20% cold worked.

For heat treatment B, all radiation hardening appears to have occurred in the first ETR exposure of 2×10^{20} nvt > 1 MeV. For heat treatments A and C, the rate of hardening was selective, depending on the ratio of principal stress. At the final ETR exposure of 7.8×10^{20} nvt, all three heat treated coupons exhibit approximately the same Knoop hardness. Also at this exposure level, hardness appears to be isotropic. The strain accompanying the Knoop hardness indent is estimated to be about 10%. The average increase in strength estimated from these Knoop hardness measurements is about 16%.

In-Reactor Corrosion of Zirconium Alloys

The November report (BNWL-CC-481-11) summarized weight gain data from the 4-cycle G-7 loop exposure of zirconium alloy corrosion specimens in Quadrants 210, 211, and 214, tested during ETR Cycles 77 through 80. Hydrogen analyses of the specimens were reported in the December report (BNWL-CC-481-12). Since the November report, selected specimens have been cleaned chemically to remove deposits remaining after the specimens were wiped clean with cotton pads. Specimens appearing to be free of deposits also were cleaned chemically to assure that undetected deposits were not present. The chemical cleaning solution was a variation of RDW-3,* containing 25 grams oxalid acid, 50 grams ammonium citrate, and 2 grams of ferric sulphate per liter of water. For general use, 1 gram of diethyl thiourea also is added to act as an inhibitor. Samples appearing clean did not undergo additional weight loss in chemical cleaning. Changes in the average weight gains from values reported in the November report are given in the following table.

*Patent designation.

In-Flux Weight Gains After Chemical Cleaning

<u>Alloy</u>	<u>Condition</u>	<u>Quadrant*</u>	<u>Wt. Gain Nov. Report</u>	<u>Wt. Gain After Cleaning</u>
Zircaloy-2	Ann - Prefilmed	210	16.1	16.2
Zircaloy-2	Ann - Prefilmed	214	44.8	44.1
Zircaloy-4	Ann - Etched	210	14.1	14.1
Zircaloy-4	Ann - Prefilmed	210	19.0	7.4
Zr-3Nb-1Sn	Quenched - Etched	210	12.9	12.8
Zr-3Nb-1Sn	Quenched - Prefilmed	210	6.7	4.4
VALLOY	Ann - Etched	210	13.7	10.9
Zr-2.5Nb	Q, 20% CW, A - Prefilmed	214	5.4	3.9
Zr-2.5Nb	Q, 30% CW, A - Etched	211	11.2	10.5
Zr-2.5Nb	Q, 30% CW, A - Prefilmed	214	9.9	9.2
Zr-2.5Nb	Ann, 20% CW - Prefilmed	210	12.3	9.9
Zr-2.5Nb	Ann, 20% CW - Prefilmed	211	10.7	10.3

*Integrated Fluxes: Q 210 1.8×10^{20} n/cm sec
 Q 211 4.3×10^{20}
 Q 214 7.8×10^{20}

Ten specimens from each quadrant and from each out-of-flux position were weighed before and after crud removal to quantify the in-reactor crud deposition during the 4-cycle exposure. Crud deposition is given in the following table, compared with results from the 2-cycle, 25.5-day exposure. The maximum G-7 loop bulk water temperature is 280 C.

Summary of Crud Deposition on Zirconium Alloys
in the ETR G-7 Loop in pH-10 NH₄OH

<u>Quadrant or Position</u>			<u>mg/dm²</u>		
	<u>Exposure</u>		<u>Maximum Deposit</u>	<u>Minimum Deposit</u>	<u>Mean Value</u>
	nvt, >1 MeV	Days			
205	2.1×10^{20}	25.5	4	0.3	2
206	3.0×10^{19}	25.5	10	0.2	4
∅1 sec*	out-of-flux	25.5	68	24	35
210	1.8×10^{20}	77.4	25	7	16
211	4.3×10^{20}	77.4	28	11	20
214	7.8×10^{20}	77.4	23.1	11	17
∅1 sec*	out-of-flux	77.4	189	41	125
∅1 min	out-of-flux	77.4	8	2	5

*Water transport times.

The in-flux quadrants and specimens ∅1 min out-of-flux were vertically mounted. The out-of-flux specimens ∅1 sec from the flux were horizontally mounted at a point where the stream makes an abrupt change of direction,

likely accounting for the relatively heavy deposit at that point. The deposit weight increases with time in-flux and out-of-flux. At the in-flux positions, deposit weight does not appear to depend on flux level.

APPLIED AND REACTOR PHYSICS

Plutonium Criticality Studies (E. D. Clayton)

Basic Criticality Studies with Plutonium Compounds in Undermoderated Range: Effect of Pu-240 on Criticality

A new series of criticality experiments were begun with PuO₂-polystyrene compacts and the remote split-table machine. The experiments are a continuation of the basic research program to provide data needed for evaluating the effects of moderation and Pu-240 in intermediate neutron spectra plutonium systems. The data are principally used for the purposes of providing an improved basis for criticality control of plutonium and its various isotopes, and for checking the computational methods on which criticality safety and reactor calculations depend. Previous experiments were with fuel having an H/Pu atomic ratio of 15 and with Pu-240 isotopic contents of 2.2 and 8 wt%. The fuel being used in the current experiments has a H/Pu atomic ratio of 5 and a Pu-240 isotopic content of about 12 wt%. A summary of results is given in the table.

For contamination control, the PuO₂-polystyrene compacts were first sealed in aluminum cans. However, this introduced a 15% void fraction into the system and, with the available fuel (≈76 kg Pu), prevented criticality from being achieved with an unreflected assembly. Furthermore, the system remained substantially subcritical even after 6.8 kg of Pu of 8 wt% Pu-240, having an H/Pu ratio of 15, were added to the assembly--increasing the length of the 12-inch rectangular prism. It is estimated that criticality would have been achieved with ≈76 kg of the 12 wt% Pu-240 and 34 kg of the 8 wt% Pu-240, with the combined critical mass of the two-core assembly being 110 kg. In order to reduce the void fraction within the assembly, the fuel cubes were then removed from their aluminum cans and, for contamination control, each cube wrapped with a 7-mil thick pressure sensitive tape capable of withstanding temperatures above 100 C. As a consequence of the energy from α-decay, temperatures up to about 80 C were reached during the course of the critical experiments.

CRITICALITY OF PLUTONIUM COMPOUNDS

Fuel: PuO_2 -polystyrene compacts, 11.5 wt% ^{240}Pu , H/Pu of 5,
230 g Pu per each 2.02 in. x 2.02 in. x 1.5 in. compact.

<u>Assembly Composition</u>	<u>H/Pu</u>	<u>Length</u>	<u>Width</u>	<u>Height</u>	<u>Critical Mass</u>
Comprised of compacts sealed in aluminum cans	5	10.7 in.	10.4 in.	8.27 in.	31.9 kg Pu
Comprised of compacts wrapped with 7 mil. thick tape.	5	10.2 in.	10.2 in.	7.75 in.	28.5 kg Pu

Phoenix Fuel Reactor Program (D. D. Lanning)MTR-Phoenix Fuel Experiment

CAF-Phoenix Fuel Experiments. One set of noise data from experiments on subcritical assemblies of Phoenix fuel in the Critical Approach Facility (CAF) has been analyzed successfully. The core assembly which contained 18 cans of fuel with 115 cells in each can, beryllium reflector, and a poison can in the center, had a break frequency of (115 ± 3) cycles/sec. This corresponds to $\frac{\beta - \rho}{\lambda}$ of (976 ± 16) .

The noise data from these experiments has proven difficult to analyze. The high impedance necessary to amplify the low level signals introduced attenuation factors from the instruments. The computer program NOISE that is used to analyze these data has been modified to compensate for instrument attenuation.

CAF-Phoenix Fuel Experiment Analysis. The analysis of the CAF-Phoenix experiments continues; the results of previously reported analyses, as well as current progress, is summarized in the following table. All of the efforts to date have been directed toward the H₂O reflected core because of the simplicity over the combined Be and H₂O reflected core. The effort of thermal spectrum mixing at the core and reflector interface was evaluated during this report period. To accomplish this objective, a mixed two-region spectrum was calculated using the integral transport theory thermalization code THERMOS. The size of this buffer cell region was determined from calculated core-reflector thermal spatial flux shapes. These calculated fluxes were from four-group diffusion theory with region spectrum averaged cross sections in the core and the reflector. The thermal flux thus calculated passed through a minimum 3 cm inside the core outer radius and went through a maximum about 3 cm into the reflector. The mixed spectrum cross section set was calculated by inserting reflecting boundaries at the positions of minimum and maximum thermal flux (zero gradient). The resulting mixed spectrum thermal cross section averages were then used in a four-region, four-group diffusion theory calculation. The four regions were:

1. Pure core spectrum, core material.
2. Mixed thermal spectrum, core material.
3. Mixed thermal spectrum, water.
4. Pure water spectrum, water.

The region radii were, of course, chosen to coincide with the position of the flux minimum, the core reflector physical boundary, the reflector maximum, and the physical boundary of the water reflector. It was found that the inclusion of the two mixed spectrum buffer zones did not appreciably change the position of the flux minimum and maximum, so that a further "iteration" on the mixed zone cell size was not necessary. The effect of the buffer zone on k_{eff} for the system was -12 mk.

Thus, spectrum mixing below 0.683 ev (the upper boundary of the thermal group) has a significant effect on k_{eff} . One would expect that the effect would be smaller for the beryllium reflector due to the less abrupt slowing-down of neutrons in the beryllium compared to the water reflector. The table following gives the relative separate effects on the calculated system k_{eff} .

<u>Source</u>	<u>$\Delta k/k$</u>
1. Variation in core composition	± 0.0013
2. Cylindricizing the hexagonal experimental geometry	$+0.0073$
3. 16 vs 3 epithermal groups	-0.0168
4. Thermal spectrum mixing at core and reflector	-0.0115

A combination of these methods will be used to analyze the Be reflected system for which k_{eff} is much better known. If the effects are additive, the previously reported 3.5% overprediction of k_{eff} would be reduced to 1.4%. The effect of multithermal groups in the whole core calculation has yet to be examined.

PRCF-Phoenix Fuel Experiment. Fabrication of all fuel and shim assemblies has been completed, and the fuel is ready for use.

Installation of the beryllium reflector in the core tank has been completed, and the mechanical structure of the PRCF-Phoenix core has been installed in the reactor cell. The safety sheet drives and flux chambers have been installed and checked. Initial fuel loading is scheduled to begin before February 1, 1967.

MTR Phoenix Fuel Fabrication. An advertisement for expressions of interest to fabricate a Phoenix fuel load for the MTR was placed in the December 15, 1966 issue of Commerce Business Daily. Expressions of interest, as a result of the advertisement, were received from Aeronca, Inc.; Nuclear Materials and Equipment Corporation (NUMEC); and United Nuclear Corporation.

MTR Phoenix Fuel Calculations. The possibility of using end fittings containing boron to reduce the very high peak-to-average power ratio (P/P) in the follower fuel below the core was investigated, and the following results were obtained:

<u>Case</u>	<u>Length of Control Rod Inserted in Core (cm)</u>	<u>N/b-cm Boron</u>	<u>Wt% Boron in Al</u>	<u>k_{eff}</u>	<u>$\frac{A}{P/P}$</u>
1	43.1	0.0	0.0	1.058	4.83
2	43.1	3.0×10^{-5}	0.02	1.039	4.23
3	43.1	3.0×10^{-4}	0.20	0.996	3.27

In cases 1 and 2, the position of the largest peak-to-average ratio remains the same, below the core in the follower fuel, and the magnitude of the ratio was decreased slightly. When the peak-to-average power ratio was determined for the case of the largest boron concentration, it was found to be considerably lower. In addition, the position of the peak was in the core just below the control rods.

Thus, poison in the end fittings will lower the peak-to-average power ratio and will move it to a position in the core in which the peak can be expected to decrease as the core is burned.

Additional refinements have been made in the control rod model and the method of representing the individual fuel elements. The control rods are simulated by a rectangular annulus containing water, aluminum, and cadmium. The width of the annulus was recently reduced to be more nearly equivalent to the thickness of the aluminum extrusion plus the aluminum clad cadmium. The amount of cadmium present in the annulus was determined by matching the known shutdown k_{eff} for an all-uranium core. The earlier or old control rod model was also in error in the geometrical representation of the uranium base or known core.

The spatial mesh within the core was modified so that the thickness of the end plates on the follower fuel and the thickness of the side plates on all the fuel was more nearly correct.

Using these refinements, a three dimensional calculation was carried out to determine the shutdown on all rods in reactivity for the 8-rod MTR core. The resulting k was 0.862.

Two dimensional calculations using the newly determined control rod model predicted a value of $k_{eff} = 0.81$ for the same configuration. Because the two dimensional calculations were four group calculations and known to predict lower values of k_{eff} , the agreement between the 2-D and the 3-D calculations was deemed acceptable.

Investigations into the effect of using borated fuel box side plates on the power peaks at the sides of the core are under way.

Phoenix Fuel Application

High Flux Isotope Reactor (HFIR) Calculations. Burnup calculations using ZODIAC-II were completed for three HFIR cores:

1. Core 1, present U-235 - B-10 load.
2. Core 2, nuclei of U-235 replaced by the same of Pu-239 and Pu-241 nuclei in a 60/24/13/3 plutonium isotope mixture, no B-10.
3. Core 3, same as core 2 except that all plutonium isotope concentrations were doubled.

The complicated fuel region involute plate geometry was represented in a calculation of thermal flux depression factors by slabs of average fuel thickness. The actual fuel plate thickness is varied within the fuel region annulus to effect some degree of spatial flux flattening. The integral transport thermalization code THERMOS was used to calculate region thermal flux averages from which the thermal self-shielding factors used in the ZODIAC-II burnup calculation were derived. Pu-240 resonance absorption in the 1_{ev} giant resonance was calculated by the NR1A approximation.

The changes in k_{eff} for the 25-day period were 9%, 8%, and 5% for cores 1, 2, and 3, respectively. The calculated region averaged group fluxes in the target zone are shown below for the three beginning-of-life cores.

Target Region Averaged Fluxes ($10^{15}m/cm^2 \text{ sec}$)*

<u>Core</u>	<u>Group 1</u>	<u>Group 2</u>	<u>Group 3</u>	<u>Group 4</u>	<u>Total</u>
1	1.143	0.655	0.084	1.662	3.544
2	1.585	0.909	0.111	1.438	4.043
3	1.534	0.918	0.094	1.033	3.579

*Energy boundaries: Group 1 10 - 1.174×10^4 (ev)
 2 1.174×10^4 - 2.38
 3 2.38 - 0.683
 4 0.683 - 0.0.

From these calculations, one would predict substantially greater life for a plutonium core in HFIR with somewhat greater total neutron flux in the target region. Of this greater total flux, a smaller percentage would be thermal flux. However, should the target material require primarily thermal neutrons, target and core design optimization could greatly increase the percentage of thermal flux available. The target region assumed in the calculations was 65 vol% water and 35 vol% aluminum. Target thermal fluxes in HFIR document, ORNL-3572, are somewhat higher than those calculated for the uranium case. The discrepancy is probably due to the crude treatment of the sophisticated HFIR reactor geometry.

PRTR-Phoenix Fuel Calculations. A series of criticality calculations were made to determine the feasibility of demonstrating a Phoenix effect in the PRTR. This study was performed using 19-rod clusters of Pu-Al with Zr cans containing various weight percents (1.8, 5, 10, 20, and 40%) of Pu with composition 60/24/13/3 percent Pu-239/Pu-240/ and Pu-242, respectively. The calculations were performed using the HRG-THERMOS-HFN codes.

These beginning-of-life calculations of the reactivity of a high power density Pu-Al core in the PRTR have shown that, in the range between 20 and 40 wt%, Pu can be loaded with little or no increase in reactivity. This behavior is necessary but not sufficient proof that a strong Phoenix effect might be expected from such a load. Accordingly, ZODIAC-II burnup calculations are under way to explore the possibility of Phoenix cores for the more thermal but high neutron economy environment of a heavy water moderated reactor.

High Temperature Reactor Lattice Physics Studies (R. E. Heineman)

Reactor Construction

The installation of the experimental core in the reactor was completed. The eight test core heater thermocouples were also installed. The thermocouples and RTD's which monitor the test core graphite temperature penetrate the reactor from the rear face hatch cover. Since the penetrations in the hatch cover are designed for a $4\text{-}3/16$ " lattice spacing, angular adaptors were fabricated to mate with the $3\text{-}3/4$ " test core lattice spacing. These adaptors were installed.

All of the reactor thermocouples (72) have been installed as has been the flux wire monitors. Blank flanges are in place over the horizontal and vertical safety rod openings in order to leak-test the shell after placement of the front door.

The nuclear safety equipment was assembled in the control room. One unit has been returned to the vendor for repair. Nuclear calibration tests have been scheduled in the PCTR in a known neutron flux. The tests are to be witnessed by the vendor who is to make any final adjustments that may be necessary.

Minor changes to the safety circuit wire runs, valve wiring, door switches, and flow switches were made and accepted. These will result in simpler and safer operation.

Seven horizontal control rods were assembled during the month. Sufficient spares exist to complete the required nine rods.

The Acceptance Test Procedure on the reactor heater control system was completed.

The gas blower was started up with the assistance of the manufacturer's representative. The clearance on the impeller was increased, and the drive and fan shafts were realigned.

Reactor Equipment

Neutron Generator. The internal parts were assembled, debugged by the vendor, successfully tested following a four-hour run, and accepted by Battelle, with minor exceptions which related only to final delivery of spare parts and prints. The generator was operated in both the horizontal and vertical positions.

Full power of 75 microamps of deuteron beam current with 2 MeV accelerating voltage on the beryllium target was maintained continuously for the four-hour test with essentially no adjustments required. This operation was well within the specification.

Gold foils were irradiated during the last hour of the test and resulted in a vertical neutron flux traverse through the reactor.

Operating and maintenance instructions were given by the vendor's representative.

Oscillators. Assembly and cold test of the oscillators is under way in the 314 Building. The light duty oscillator is scheduled to be ready for cold testing of the drive system and positional readout at month-end.

The light duty oscillator valve is completely assembled, has been tested, and is ready for installation on the reactor. The heavy duty oscillator valve is assembled and has been tested for leakage across the valve plate and inflatable seal and has met requirements for this portion of the test. Test of the valve for leakage as a whole unit has not been satisfactory. Faults are currently being corrected, and the entire valve will be retested.

The keys for attaching the oscillator ram to the test sample are complete and are being fitted to the light duty oscillator. The logic system for position readout has been modified to accommodate adjustable acceleration and deceleration ramps and to permit continuous oscillation.

It is estimated that the oscillators are 80% complete.

The HTLTR mockup is being prepared for hot tests of the Light Duty Oscillator system. The door of the mockup is being modified to receive the oscillator valve plate and valve seal.

The test will use a three-inch hole in the test block; a three-inch diameter test piece weighing 1.5 times the design specifications will be inserted, oscillated, and removed at temperature; three heater rods will be used in the mockup instead of four, which will allow a test of the rods to essentially full power for sustained periods at temperature; a few samples will be inserted into the mockup. These include copper and gold foils, fuel element as assembled, a gadolinium shim rod as received, and either a safety blade or sample safety blade material.

Control Rods. The manufacturer of the motor transducer units was asked to evaluate the design of a rod drive unit with particular emphasis on the reliability and instability of the stepping motor feedback circuit. After review of the design, the manufacturer believed that the design was an excellent example of a good design using his product and suggested that the position readout test equipment be changed. He believed the trouble was in the test and not the drive unit. Following this suggestion, the feedback problem on the control rod drive was solved.

Programmed Measurement and Control System. A limitation was discovered in the Flux Digitizer for PMACS. It was found that at least one computer cycle must elapse between the time a transfer is requested from the flux accumulator to the flux storage register and the time a transfer is requested from the flux storage register to the computer accumulator. This time delay is necessary to allow the flux storage register to become set.

A new printed circuit character decode card was designed and built for the 3-color alpha-numeric display. This new character card allows two characters to be decoded on each card, doubling the character capacity of the character generator. A "minus sign" and a "period" was added to the display capability using the new character card. Additional printed circuits and components have been ordered to allow more characters to be added.

Reactor Operations

The systems design tests are being rewritten to make use of the programs written for the computer. The rough draft is now 90% complete. These tests bear a close relationship to the Operating Procedures which are about half finished. Work on valve manuals and operator qualification tests continued.

A Criticality Safety Specification for the storage and handling of the control rods (which contain enriched uranium) has been written and is being circulated for approval.

Eight field modifications were made to the PMACS PDP-7 computer by a Digital Equipment Corporation representative. Such field changes are made periodically by the manufacturer to update and to improve the performance of the computer.

The stored programs controlling the gas flow in the reactor were expanded to include operation at all temperatures. Flow charts have been completed for these procedures. The gas system consists of nine subprograms that correspond to the nine possible operating states for the gas system. The states may be entered at the command of the operator or automatically under program control. The nine subprograms are:

Low Temperature Gas Control

Evacuate
Fill
Recirculate
Vent
Shutdown

High Temperature Gas Control

Heat
Cool
Standby
Emergency

The proposed Operating Specifications were reviewed, and a few changes were suggested.

Reactor Physics Program

Startup and Calibration Experiments. During the acceptance test operation of the neutron generator, a number of gold foils were inserted in the moderator block. Analysis of the results indicates that the neutron distribution is approximately $\phi = 3.46 \times 10^6 e^{-.023Z}$, where Z is the vertical distance above the bottom of the graphite in centimeters. Although the accuracy is poor, the inferred diffusion length for the graphite is about 55 cm.

Oscillator Supports and Couplings to Experimental Core. Preliminary designs for the oscillator supports and couplings to be used in the tests in the 314 Building have been conceived. The design will be finished by the Equipment Development group and construction started shortly. The results of the tests will indicate the final design to be used in the HTLTR.

Fuel Element Heater. Design of a fuel element heater has begun. The heated fuel element will provide the fuel temperature coefficient for the startup experiments. This fuel element will be designed to reach 500 C. The $U^{233}O_2$ -ThO₂ heated element is expected to be more complicated, although some of the design features of the startup element may be pertinent. The $U^{233}O_2$ -ThO₂ element will be designed to reach temperatures of 1200 C.

$U^{233}O_2$ -ThO₂ Experiments. Planning for the first experiment was continued. Calculations using GAMTEC and THERMOS have been started to predict the experimental quantities to be measured.

Evaluation of Temperature Dependent Reactor Calculations. For comparison and calibration purposes the initial chain reacting media to be tested in the HTLTR is a 1.36-inch natural uranium rod in a 7.5-inch graphite lattice. This configuration was previously investigated by Lloyd, Clayton, and Richey (HW-51008) in a heated exponential measurement. As a first attempt to evaluate current theoretical methods for computing temperature coefficients, a study was conducted for the HTLTR startup experiment using calculational techniques recently developed. It is recognized that although current techniques may accurately predict the very integral exponential experiments, the calculations undoubtedly have compensating errors.

The calculational methods consisted of the usual two-group critical expression, i.e.,

$$1 = \frac{(\eta f)_{Fast}(1 - p)}{1 + B_C^2 \tau} + \frac{(\eta f)_{Th} p}{(1 + B_C^2 \tau)(1 + B_C^2 L^2)}$$

and $k_{\infty} = (\eta f)_F(1 - p) + (\eta f)_{Th} p.$

Each parameter in the above expressions was obtained using the GAMTEC-II code. This code computes the parameters from an equivalently homogenized unit lattice cell with: (1) corrections for fuel lumping on the fast fission of U-238, (2) application of NR and NRIA approximations in computing resonance capture in U-238, and (3) P_3 computed flux for a flux X volume homogenization at thermal neutron energies. The thermal neutron spectrum was obtained with the Wilkins heavy gas model. The computed coefficient is

$$\frac{1}{k_{\infty}} \frac{\Delta k_{\infty}}{\Delta T} = -4.1 \times 10^{-5}/^{\circ}\text{C}$$

and compares with a value derived from the referenced exponential experiment of

$$\frac{1}{k_{\infty}} \frac{\Delta k_{\infty}}{\Delta T} = -4.6 \times 10^{-5}/^{\circ}\text{C}$$

Utilizing the THERMOS code with Park's graphite scattering kernels for thermal neutrons yields a coefficient of

$$\frac{1}{k_{\infty}} \frac{\Delta k_{\infty}}{\Delta T} = -3.4 \times 10^{-5}/^{\circ}\text{C}$$

This latter result was computed with more accurate technique for the important thermal group, thus its poor comparison with derived experimental data indicates the magnitude of compensating errors.

Driver and Flux Adjusting Fuel Rods. Assembly of the driver fuel rods and flux adjusting fuel rods was completed. The driver elements consist of approximately 1575 kilograms of 5 wt% enriched UO_2 pellets (1.06" dia.) clad in graphite. The flux adjusting rods (120) consist of normal UO_2 pellets (0.5" dia.) clad in graphite. Aluminum oxide pellets were inserted into one end cap of each flux adjusting rod as an identification mark. Additional identification markers having a hexagonal, square, pentagonal, and triangular shape were machined for the Gd poison rods and for future experimental fuel rods.

Safety Analysis Report. Discussions were held with A. J. Presseky, AEC Headquarters, to evaluate questions raised by various personnel in RDT. The questions were answered orally, and a written reply is now in rough draft. Some changes in wording of the SAR and additional figures will be supplied in the near future.

Data was also generated in answer to a question concerning the movement of the central experimental core, either deliberate or during an earthquake. It was calculated that the reactivity addition which could be caused by movement of the central core during an earthquake would be less than \$1.00. Deliberate insertion of a fueled test core, without removal of driver fuel, amounts to the same consequences as a fuel loading error.

REACTOR FUELS AND MATERIALS

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

Hot Pressing of UN-PuN

Work has been initiated to determine conditions necessary for hot pressing UN - 20 wt% PuN pellets to >95% TD for irradiation testing. Previous work at other sites indicates that both UN and UN - 20 wt% PuN require pressure-temperature conditions of 10,000 psi at 1500 C to achieve this result.

The concensus of vendors and users* of zirconia and alumina hot press dies is that neither of these materials is suitable for use at the required temperature-pressure conditions. Apparently only graphite or TaC can be used in this application.

UN-PuN Synthesis

The mixed nitride production facility has attained "nearly routine" operation. Each of the unit operations involved has been demonstrated to be operable. Preliminary runs indicate that the facility is limited to the production of approximately 250 grams of mixed nitride pellets per week. The status of each of the unit operations is as follows:

PuN Synthesis. Two PuN synthesis runs have demonstrated that approximately 20 grams of material can be successfully prepared in a batch which requires two days to complete. The synthesis technique being used consists of hydriding Pu metal at room temperature, then reacting the PuH with N₂ at approximately 800 C to form various nitrides of Pu. These higher nitrides are reduced to PuN by continued heating at approximately 1000 C.

UN Synthesis. Three runs made in the UN facility revealed some processing problems, but these have been successfully resolved so that batches of approximately 150 grams each can now be made in two days elapsed time. The synthesis technique being used is very similar to that for PuN. Uranium metal is hydrided, dehydrided, nitrided, then reduced to form a high purity product. The first two runs made were unsuccessful because hydriding and subsequent nitriding of the uranium metal were incomplete. This was corrected when it was realized that the total system volume was too small to contain enough hydrogen to complete the hydriding step. Makeup hydrogen is now continually added to the system until the hydriding reaction is complete. A similar procedure is followed during nitriding. The third synthesis run, in which this technique was used, produced apparently satisfactory material.

* Coors Porcelain Co., Golden, Colorado; Zircoa, Solon, Ohio; Sandia Corp., Albuquerque, New Mexico.

Analysis of UO₂-PuO₂ Fuel Cycles for Fast Reactors

A review of existing work is nearly complete. Preliminary coding and running of test cases for debug purposes will be finished during February.

Basic Swelling Studies (R. D. Leggett)

The purpose of this program is to characterize and understand the behavior of fissionable materials during irradiation. The theories developed provide a basis for the engineering exploitation of metallic fuel materials in advanced reactor applications. Specimens under study at the present time include uranium with and without dilute alloy additions.

Irradiation Program

Capsule 36 (450 C, ~30 psi) was discharged after operating successfully to an estimated exposure of ~0.35 at.% BU. Capsule P-15 (700 C, 1000 psi) was charged into a reactor and will be irradiated to 0.2 at.% BU in the natural uranium specimens. Capsule P-15 contains four levels of U-235 which will provide different burnups within approximately the same environment to enhance the evaluation of the effects of burnup and burnup rate.

Construction is almost complete on two controlled temperature-pressure capsules (P-14 and P-16). Capsule P-14 (700 C, 500 psi, 0.2 at.% BU) is ~2/3 complete, and capsule P-16 (625 C, 1000 psi, 0.35 at.% BU) is in the final stages of preirradiation testing.

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

Postirradiation Examination

The electron microscope examination of uranium specimens postirradiation annealed at 625 C for 100 hours indicated that no change occurred in the as-irradiated structure. Both specimens had been irradiated to 0.2 at.% BU; one at 610 C (alpha phase) and the other at 690 C (beta phase). The specimen irradiated in the alpha phase still shows grain boundary porosity and a small amount of matrix porosity. The specimen irradiated in the beta phase contains cracks (due to the transformation from beta to alpha) and matrix porosity of about the same size and distribution that existed prior to annealing. Both specimens contained a large number of small, unidentified second phase particles, part of which are suspected of being artifacts. The size of the particles appeared to be somewhat larger in the specimen irradiated in the beta phase.

High Pressure, Postirradiation Annealing

The modification and development of equipment and facilities to post-irradiation annealed uranium to 15,000 psi and 900 C are partially completed. The necessary apparatus for in-cell welding was obtained and installed, and excellent welds were made remotely on sample capsules. After the capsules are loaded with irradiated uranium and the first closure weld made, they will be transferred in a cask to a glovebox for NaK filling and the final seal weld.

Numerous tests were made of the cold wall autoclave at increasing pressures up to ~7000 psi while trying to maintain approximately 900 C in the test zone. Various changes in insulation and baffle arrangement have cut down on convection currents and have reduced heat losses, but the existing heater wattage in certain zones is inadequate to maintain the desired temperature at the higher pressures. During the final test with a new baffle arrangement, the bottom heating zone apparently shorted out. It is suspected that moisture contained in the as-received microquartz insulation condensed at the cool bottom and caused the electrical short.

The autoclave is now being removed to install the original heater which has greater heating capacity but which requires some repair. All new thermocouples will also be installed.

Nondestructive Testing (J. C. Spanner)

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

Investigation of critical angle ultrasonic techniques as a means for detecting irradiation induced shifts in the nil-ductility-transition temperature of steel is continuing. These methods have been demonstrated capable of distinguishing neutron irradiated low-carbon steel samples from nonirradiated samples, although response anomalies were found to exist in both the irradiated and nonirradiated samples. Experimental methods are being developed whereby response anomalies will be minimized.

An alternate specimen design is being considered since gamma heating within the earlier design would have raised specimen temperature considerably beyond the 550-600 F desired. It now appears unlikely that test specimens can be ready for reactor loading by February 15; therefore, an alternate reactor position, with a somewhat later loading deadline, is being considered. Ambient temperature at the alternate position is about 140 F as compared to 550 F in the G-7 loop. Preliminary studies indicate that by varying the specimen-to-capsule helium gaps, it may be possible to maintain a basic high temperature for one group of specimens and a basic low temperature for a second group, despite an approximate 10:1 variation in gamma heating. Temperature control is an important facet in specimen irradiations because of (1) potential in-reactor annealing of irradiation damage, (2) possible intermetallic diffusion between specimen and capsule, and (3) possible phase transformations or grain growth. Any of these temperature related effects could in turn affect the

ultrasonic response. To minimize thermal effects, preirradiation heat treatments are being studied; furthermore, the in-reactor thermal history (at the specimen surface within the capsule) must be calculated and/or measured so that a similar heat treatment can be given the unirradiated control specimens.

Millwright and pipefitting work on the specimens manipulating facility has been completed. Some electrical work as well as certain mechanical modifications remains to be done. Four electronic instrumentation chassis have now been fabricated and received.

Eddy Current Probe Design Studies

A study has been initiated to investigate the theoretical and practical aspects of eddy current sensing probe design and to apply this information to the development of general guidelines for future probe construction.

Reduction of the general goal to specific identifiable steps or tasks requires careful consideration due to the large number of variables and the complex manner in which they are interrelated. It is apparent that some simplifying assumptions will have to be made; however, the initial analysis is being kept as broad as necessary to give at least preliminary consideration to all known variables.

Each testing situation is characterized by a number of test conditions such as: (1) operating frequency, (2) general coil configuration or type, (3) absolute or differential operation, and (4) physical characteristics of the probe coil or coils. The first three test conditions are generally dictated by: the characteristics of the test specimen, and the properties to be measured or detected. (Most of the design criteria for determining these test conditions have been widely known for many years.) The design criteria for the selection of probe coils, on the other hand, is not well understood, and very little published information exists.

The decisions made in specifying the test conditions, as outlined above, also determine the operational characteristics of the test system such as sensitivity, stability, area of search, etc. The problems which must be solved are therefore: (1) determine the relationships between test conditions and operational characteristics, and (2) control the test conditions to achieve the desired operational characteristics such as increased sensitivity and stability.

In order to better understand the electrical characteristics of eddy current probes and to relate them to the operational characteristics of the test systems, six possible equivalent circuits have been developed, and the theoretical complex impedance of these circuits is being determined as a function of frequency. Simultaneously, the complex impedance of several eddy current probes will be experimentally measured as a function of frequency. The information obtained from these data will then be compared with the theoretical work to establish the most representative equivalent circuit or circuits.

Fundamental Ultrasonic Studies

Analytical ultrasonic wave propagation studies using Kautz functions continued. Further investigation of a dual frequency system for detecting unbonds has shown that the signals received from areas suspected of being unbonded were actually caused from surface imperfections in the fuel element cladding.

An EDPM program was written which will evaluate the frequencies, decay rates, and relative amplitudes of the first ten pairs of ortho-normal Kautz sequences present in a given time sequence (ultrasonic pulse). This program replaces the program based on a Fourier series analysis. The use of the Kautz analysis in the main ultrasonic wave program required two subroutines to also be written: one to fit the functions in the frequency domain, and the other for evaluating points on the fitted functions in both the time and frequency domains. This work is now about 1/3 complete.

As discussed in earlier reports, the detection of unbonds at interfaces between acoustically dissimilar materials has always been a problem when the two materials are held in tight mechanical contact. Attempts were made to: (1) momentarily "open" such an interface in a reactor fuel cladding with a high power, low frequency ultrasonic beam, and (2) simultaneously transmit and reflect a high frequency beam from the same location. It was hoped that the high frequency beam would then be reflected with improved sensitivity to the unbond during the time that the interface was opened.

Preliminary experiments showed apparent success, but it was subsequently shown that the test was primarily sensitive to surface conditions in the fuel cladding and not to the suspected unbond. Additional work is being undertaken to determine if the sensitivity to surface conditions can be diminished.

Electromagnetic Testing

Additional data were obtained for use in the study relating single frequency, multifrequency, and pulsed eddy current nondestructive test techniques. Test coil output signals were measured for several different test conditions. Improved measurements were made showing the relative effects of varying test coil diameter and test frequency by using a lower range of test frequencies to reduce parasitic loading effects of unused test coils and by making physical changes in the test arrangement to improve its electrical stability.

Data on test coil outputs using pulsed excitation are being obtained using a pulsed current source for driving the test coil, a signal amplifier using both active and passive filters, and a cathode ray oscilloscope for displaying the output of the amplifier. Measurements have been made showing the effect of R-C filters, C-R ladder networks and L-R ladder networks on test coil output signals for variations in test specimen thickness and depth of defect.

The range of test frequencies in the multifrequency measurements was lowered to 2.5, 5, and 10 kHz in contrast to the original 25, 50, and 100 kHz in order to reduce the parasitic loading effect of unused test coils. The test coil assembly consists of ten concentric, coplanar coil pairs ranging from 1/8" diameter to 1-1/4" diameter in 1/8" steps, mounted in 10 circular slots in a plexiglass sheet 4" x 1/4". Each coil pair consists of a 10-turn primary winding and a 10-turn secondary winding. The present measurements are made using in turn the 1/4", 1/2", and 1" coil pairs. When one coil pair is being used, all other coil pairs are open circuited. However, it was observed that at the higher test frequencies the unused coils electrically loaded the coils being used, resulting in a shift of test coil test specimen conductivity locus in the phase lagging direction. It was found that the degree of this effect varies with frequency, and measurements show that it is due mainly to eddy current losses in the unused coils. The behavior of the effect is further complicated due to variation in coupling between the coils and variation in distributed capacitance as a function of position of the test coil assembly with respect to the test specimens. This effect was reduced to acceptable levels by lowering the frequency range used.

The measurements confirm previous results which showed that the test coil output signals in test specimen electrical conductivity tests varied as test frequency was increased in a manner similar to that observed when the test coil diameter was increased. The test results also show that these effects vary in a complex way depending upon frequency, coil diameter, and test specimen conditions, and further study is needed. However, the degree of independence of signals from coils having diameters equal to 1/4", 1/2", and 1", all operating at one frequency, seems sufficient to permit operation of a multidimensional thickness or conductivity test using only one test frequency.

Infrared and Thermal Research

Performance tests on the sinusoidal thermal wave instrument during an eight-week period have shown that its long term stability is not yet within acceptable limits. Daily standardization, proposed earlier, would not eliminate the problem since the shift in calibration was not the same for all samples. A study to determine and correct the causes of the instability is under way. Thus far, intermittent difficulties have been found in the amplitude digitizer and the phase printer, and other components that effect the phase angle were found to be temperature sensitive.

Work is continuing on a remote transient method for measuring surface thermal impedance values for various materials. The method makes use of a high intensity xenon flash lamp to irradiate the sample surface. The resulting temperature transient is measured with a sensitive infrared radiometer. The Laplace transform of the transient should then yield the surface thermal impedance directly.

A trigger circuit to trigger the flash lamp at the start of sine and cosine waves, required to electronically evaluate the transform, is being fabricated. A function generator to supply both fixed and variable phase sine and square wave outputs has been ordered. The fixed phase square wave is used to trigger a silicon controlled rectifier which in turn discharges a capacitor into the trigger transformer primary. The resulting high voltage pulse at the secondary is used to trigger the flash lamp.

A mathematical analysis has been performed to investigate the effects or the finite bandwidth restrictions of the radiometer system on the phase angle of the measured surface thermal impedance. It was reported earlier that phase angles of -53° for plate glass and teflon and -75° for copper and stainless steel were measured using a graphical technique to evaluate the Laplace transform of the temperature transient. It was thus of interest to determine whether or not the bandwidth restrictions mentioned above contributed to errors in the phase. (Theoretically, the surface thermal impedance should have a phase angle of -45° for an impulsive heat source.) The calculations indicate that the bandwidth restriction introduces no appreciable error provided $WRC \ll 1$, where W = angular velocity of the test frequency and RC = the resistance x capacitance product for a simple low pass filter which has a break point at the upper frequency limit of the radiometer pass band (the bandwidth is d.c. to f_b , the break-point frequency).

Nuclear Ceramics (R. E. Nightingale)

Materials and Information Exchange

Single-crystal specimens of ThO_2 and UO_2 were prepared, characterized, and sent to General Electric Research and Development Center for their use in basic studies. High purity ThO_2 and $\text{ThO}_2 - 4 \text{ wt\% } \text{UO}_2$ pellets were prepared and sent to the Bell Telephone Laboratory (Murray Hill) for their use in far-infrared studies. A cylindrical, polycrystalline UO_2 specimen was prepared and sent to Oak Ridge National Laboratory for in-pile thermal conductivity studies. Pneumatically-impacted UO_2 specimens were selected and sent to AECL, Chalk River, Canada, for use in comparison thinning studies. In addition, UO_2 pellets were selected, tested, and sent to Battelle-Northwest's Chemistry Department for use in dissolution and decontamination studies. Current requests for information and materials include: UO_2 crystals for ANL, ORNL, Pennsylvania State University; and PuO_2 specimens for ANL, ORNL, and Sandia.

The US/Japan research newsletter number 5 was compiled and prepared for printing for exchange of information of the cooperating agencies in the United States and Japan.

Thermal Diffusivity of UO₂-PuO₂

The thermal diffusivity of (U_{0.8}, Pu_{0.2})O₂ was measured from 100 to 1500 C using a laser-pulse thermal diffusivity apparatus. Thermal conductivity values were estimated from the diffusivity data and heat capacity of UO₂. The calculated thermal conductivity of the mixed oxide composition for all temperatures was within 10% of that reported for UO₂ by Godfrey et al.⁽¹⁾

Samples were prepared from coprecipitated (U_{0.8}, Pu_{0.2})O₂ powder which was densified by pneumatic impaction to 10.6 g/cm³ (95.5% TD). The oxygen content of the sample was controlled at the stoichiometric composition during diffusivity measurements by heating in a flowing atmosphere of argon - 8% H₂ passed over 0°C water.

Pu-O-N Ternary Equilibria

A ternary evaporator for depositing compositions corresponding to isothermal sections of the Pu-O-N pseudo-ternary phase diagram onto platinum substrates was assembled and is being calibrated. Tentative operating conditions call for a pressure of plutonium of 10 microns, which occurs at a temperature of 1500 C. This requires oxygen and nitrogen flow rates of about 0.01 cm³/sec and a vacuum system pressure of less than 10 microns. All of these conditions can be achieved. The platinum substrate will be held at about 800 C for the first experiments.

High purity cerium metal will be used as a stand-in for plutonium until the apparatus has been proven. Initial experiments are under way to determine the evaporation behavior of the cerium. Following these experiments, deposition of Ce-O-N sections will be attempted. The apparatus will then be located in a hood for Pu-O-N experiments.

High Temperature Electrical Conductivity of UO₂

Computer analysis showed that the electrical conductivity of UO₂ above 1400 K can be represented by the equation

$$\sigma = 2.104 \times 10^{-2} T^{1.401} e^{\frac{-0.916 \text{ eV}}{kT}} \text{ mho/cm}$$

This equation is nearly identical to the theoretical expression for intrinsic conductivity, where

$$\sigma \propto T^{3/2} e^{-E_g/2 kT}$$

The $T^{3/2}$ term is related to an increase in the number of carriers.

Previous analyses, which did not consider the $T^{3/2}$ term, gave an activation energy of 1.15 eV.

(1) T. G. Godfrey, W. Fulkerson, T. G. Kollie, J. P. Moore, and D. L. McElroy, "Thermal Conductivity of Uranium Dioxide and Armco Iron by an Improved Radial Heat Flow Technique," ORNL-3556 (1964).

Electron Microscopy of Mixed Oxides

Two specimens of sintered mixed UO_2 - 20 wt% PuO_2 , one mechanically mixed and one coprecipitated, were examined by optical and reflection electron microscopy. Both materials displayed marked porosity. The mechanically mixed specimen contained an unidentified white lenticular phase, approximately 10 microns in length, which was removed by etching. The coprecipitated material contained a plate-like white phase of similar size was retained after etching. Some of the grain boundaries in the as-polished, mechanically-mixed specimen were evident by electron microscopy, whereas the coprecipitated oxides required etching to reveal the small grains. Preparations have been completed for electron microscopy of these specimens during heating to about 800 C.

Specimens of these materials and of three other uranium-plutonium oxide samples (two sintered and one pneumatically impacted) are being mechanically thinned for transmission microscopy. The first optical micrographs show some differences between the impacted and sintered specimens, but preparation methods may be partially responsible. Development of the thinning technique is continuing.

Nuclear Graphite (R. E. Nightingale)

High Temperature Graphite Irradiations

The high temperature graphite capsule, GEH-13-13, is now operating in the ETR. Unfortunately, the capsule is adjacent to a control rod which depresses the flux; therefore, the capsule is presently operating at about 1200 C, which is 150 C lower than designed. Withdrawal of the control rod during the course of the cycle will shift the flux upward, resulting in considerable temperature variation within the capsule; however, the withdrawal should increase over-all temperatures.

At startup, the desired maximum operating temperature of 1400 C was attained easily with an equal mixture of helium and nitrogen. As time progressed, however, the temperature continually decreased. This is probably due to blackening of the internal capsule wall, causing greater heat loss through thermal radiation; this effect has been observed before following startups, but not to such an extent. If operating temperatures cannot be increased, control will be maintained at a maximum temperature of 1250 C.

All but one thermocouple (lost during initial startup) are operating well. A tungsten-rhenium and Geminol thermocouple imbedded in the same sample holder agree within 10 C.

EBR-II Graphite Irradiations

Two additional pins containing graphite have been included in 19-pin sub-assemblies in Row 6. They are scheduled to remain in the reactor for

approximately one and two years, accumulating about 3×10^{22} and 6×10^{22} nvt ($E > 0.18$ MeV) exposure, respectively. The maximum irradiation temperature will be about 550 C. Samples included in these capsules are those which have shown good dimensional stability at lower exposures. Data from these high exposure pins will extend results from earlier EBR-II graphite irradiations.

The joint graphite irradiation tests by ORNL and BNW have been tentatively approved for a Row 7 position. The flux intensities in this position will generate a maximum of 10^{22} nvt per year. The larger pins (0.806" dia.) of the 7-pin subassembly to be utilized in these irradiations will permit the attainment of higher operating temperatures than would be possible with the smaller pins of the 19-pin subassembly.

Irradiation of Nuclear Graphite

The nuclear graphite irradiation tests continue to operate satisfactorily. Capsule H-3-13 was removed from GETR on December 27. All nine thermocouples functioned properly for the complete four-cycle irradiation.

The exterior surface of the capsule showed no unusual wear, and the internal components were in the proper locations and intact. No samples were damaged or broken.

At the maximum exposure of 2.38×10^{22} nvt ($E > 0.18$ MeV), the NC-8 graphite irradiated at 800 C displays net length changes of +19.8% and -7.5% in the transverse and parallel directions, respectively; the corresponding volume change is +33%. The CSF material, at the same temperature and exposure displays net length changes of +10.1 and -7.8% in the transverse and parallel directions, respectively; the corresponding volume change is +11.7%. On both materials a decreasing contraction rate for the parallel orientation indicates a possible "turn-around" at this exposure and temperature.

The next capsule in this series, H-3-14, is being assembled and will be charged into the D7 position of the GETR the first week in February.

Irradiation of "Dimensionally Stable" Graphite

The H-3-24 capsule, in its third GETR cycle, continues to run at design temperature. Due to problems with reactor-associated equipment, the GETR is now operating at a reduced power level of 40 MW. Full power operation is expected in March or April. Five of the nine thermocouples have failed; however, by comparing temperature readings from the four remaining thermocouples with temperature profiles from past cycles, the temperature distribution along the capsule length can be deduced.

Irradiation of Pyrolytic and Single Crystal Graphite

The irradiation of eight capsules of pyrolytic and single crystal graphite from Union Carbide Corporation, Carbon Products Division, has been completed. Exposures on the capsules remain to be determined. Four capsules were irradiated at 50 C, three at 650 C, and one at 1000 C. All samples from the irradiations are being returned to Union Carbide.

Averaging Techniques in Polycrystalline Graphite

In previous monthly reports the work on averaging techniques in polycrystalline graphite has dealt with the effect of c-axis accommodation and crystallite orientation on Young's moduli and thermal expansion coefficients for the limiting approximations of constant stress and constant strain. Although the limiting approximations of constant stress in conjunction with accommodation will give reasonable values of thermal expansion coefficients and Young's moduli, the pure constant stress model is inadequate when porosity or the c-axis accommodation mechanism is considered.

Two compromise models have been developed to obtain reasonable answers from a more realistic model. The first model, referred to as a modified constant strain model, has previously been used by Buessem to explain the properties of some polycrystalline anisotropic ceramic materials. (See the November 1966 monthly report.) The alternative model will be called a "modified constant stress model" and consists of a constant stress average of crystallite pairs which are formed by connecting two crystallites in parallel as in the constant strain approximation. It has been formulated to introduce a mechanism for c-axis accommodation into the constant stress model.

A numerical evaluation of these modified models for isotropic graphite gives very good agreement with the experimental results. Values of Young's moduli for these two models fall between the two limits of constant stress and constant strain as required by energy considerations. However, they lie very close to the value predicted by the constant stress model rather than the average value of the two limits. Similarly, for the thermal expansion coefficients the predicted coefficient falls between values calculated from the two limiting models. Based on this agreement, these models represent good approximations to the structure of polycrystalline graphite and should be useful in predicting or explaining other properties.

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

Alloy Selection

Fabrication of the program heat of Incoloy 800 is essentially complete. A more favorable schedule for cold drawing of the tube blanks to final size was negotiated with Huntington Alloy Products, Inc. As a result, the tube blanks are undergoing ultrasonic and final surface inspection on January 26 and 27, 1967. Sheet stock on this order will be furnished in two thicknesses and two conditions as follows:

5 pcs., 0.109" x 12" x 48", solution annealed;
5 pcs., 0.083" x 12" x 48", cold-rolled, annealed.

The liquid sodium, irradiation capsule, GEH-22-4, will soon complete 90 full power days in the ETR with an estimated total fast fluence of 1.3×10^{21} nvt. The capsule will be discharged at shutdown Cycle 87, about January 30, 1967. There is evidence that the gas outlet line has recently become plugged. Possible causes for this happening are now being investigated. Temperature readings, from the remaining thermocouple, indicate nothing abnormal in capsule function. The cause of the line plugging should be revealed during postirradiation examination.

The liquid metal capsule irradiation program is being shifted from the M7 to the G6 core position in the ETR. Examination of the core characteristics of the G6 position reveals possibly superior flux and gamma heating characteristics compared with M7 position.

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 and nickel-base alloys during neutron irradiation.

A strain versus time curve was obtained for an in-reactor creep test conducted on Hastelloy X-280 at 1000 C temperature and 3500 psi stress. The data show very little first-stage creep. The total elongation was greater than 11% as measured by the in-reactor extensometer system. This value will be verified by postirradiation specimen examination. Rupture occurred after about 75 hours of actual test time.

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.

AISI 304 stainless steel tensile specimens, irradiated and tested at various temperatures, have been sectioned for electron microscopy. Microscopic examinations have been initiated with the intent of characterizing the nature of the irradiation-induced damage in this alloy. Electron microscopy on AISI 304 stainless steel creep samples is still in progress.

Capsules have been designed for conducting irradiations at temperatures of between 400 and 600 C in the ETR G-7 loop.

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 preirradiation thermal or thermomechanical treatments and are evaluated by tensile tests, stress-rupture tests, and microstructural examinations.

Stress-rupture tests at 1350 F (735 C) were conducted on unirradiated, pretreated Hastelloy X-280 specimens that were controls for irradiated specimens tested previously. Preliminary data reveal a retained beneficial effect of the pretreatments on rupture life.

Stress-rupture tests at 1350 F (735 C) and 9800 psi (the unirradiated, 1000-hr rupture stress) are nearing completion on eight, pretreated, Inconel 600 specimens which were irradiated at 540 F (280 C) to an approximate fluence of 1×10^{20} nvt ($E > 1$ MeV). Results of these tests will be compared to similar tests conducted on Hastelloy X-280. Also, stress-rupture tests at 1350 F (735 C) and 37,500 psi (the unirradiated, 1000-hr rupture stress) have been initiated on pretreated, Inconel X-750 specimens which were irradiated at 540 F (280 C) to an approximate fluence of 1×10^{20} nvt ($E > 1$ MeV).

Tensile specimens of Inconel X-750, representing several new pre-irradiation treatments designed to improve high temperature postirradiation ductility, are being irradiated at 540 F (280 C) to a goal fluence of 1×10^{20} nvt (> 1 MeV).

SAP Alloys. A program to determine the effects of irradiation on Sintered Aluminum Products (SAP) in support of the HWOCR project is being initiated. Sample quantities of XAP 005 and SAP 895 rods were received from Atomics International (AI) for evaluation. Preliminary characterization of these materials, to include metallographic examination and mechanical property determinations, is in progress. The tensile, creep-rupture, and notched creep-rupture properties will be determined after specimen designs have been evaluated. Sheath capsules have been designed for irradiating SAP materials in the G-7 loop at 575 and 750 F (300 and 400 C). The temperatures of 575 and 750 F, which are above the 540 F (280 C) bulk loop temperature, will be obtained by gamma heating and a helium gas gap within the capsule. The prototype capsules being fabricated will be placed in loop with thermal monitors to determine whether or not the designed gas gaps are adequate to obtain the desired irradiation temperatures.

Irradiations will also be conducted in instrumented, temperature-controlled capsules provided by AI. Capsule designs have been reviewed by PNL personnel and suggested improvements have been discussed with AI personnel. Review of existing ETR irradiation program authorizations between PNL and Idaho Nuclear Corporation reveals that the AI capsules can be irradiated under the GEH-14 irradiation program.

In-Flux Oxidation of Titanium, Niobium, Niobium-1% Zirconium, and Zircaloy-2. Titanium (Grade II), niobium, and niobium-1 wt% zirconium (3 specimens each) were exposed in the G-7 loop of the ETR in Quadrant 217. Specimens of zirconium alloys were exposed in the same quadrant. The specimens were tested in the form of one-inch square coupons; niobium specimens were 0.062 inch thick, and titanium specimens were 0.050 inch thick. The test conditions are summarized in the following table.

G-7 Loop Exposure Conditions - Quadrant 217

Time at temperature	37 days
ETR Cycles	78 and 79
Time at full power	44 days
Nominal loop pressure in sample section	2150 psi
Flux	5.9×10^{13} nv, >1 MeV
Fluence	2.2×10^{20} nvt, >1 MeV
Water Chemistry:	
Coolant	pH-10 NH_4OH
Oxygen, ppm	<0.05
Hydrogen, cc/kg	45*
NH_3 , ppm	9.1*
NO_2 , ppm	0.01*
NO_3 , ppm	<0.05*
Cs-138, dis/min-ml	4×10^4 *

*Mean values.

Out-of-reactor control specimens were exposed in an autoclave at Battelle-Northwest operated at 280 C with pH-10 NH_4OH . In-reactor and out-of-reactor weight gains for the three types of specimens appear below. The weight gain for Zircaloy-2 specimens in the same quadrant is included for comparison.

Gold interference colors appeared on in-flux and out-of-reactor titanium specimens. Weight gains were small in both environments. While the average of the three in-flux weight gains is higher than the out-of-flux weight by a factor of ~ 4 , the difference must be considered within the range of experimental error until the in-flux specimens have been weighed on a microbalance.

In-flux weight gains on niobium and niobium-1% zirconium specimens were highly variable, suggesting that sluffing occurred. Klopp, Berry, and Maykuth (Columbium Metallurgy, p. 689) report high weight gains and subsequent weight losses on pure niobium at 316 C. The surface was reddish-brown after the in-flux exposure. The out-of-flux oxide was black. Cleaning the in-flux specimens in 10 wt% oxalic acid solution resulted in removal of the reddish-brown film, resulting in a silvery surface. The oxalic acid treatment resulted in net weight losses.

In-Flux and Out-of-Reactor Weight Gains and Hydrogen Pickups
at 280 C in pH-10 NH₄OH

<u>Specimen</u>	<u>37-Day Weight Gains</u>		<u>Hydrogen Absorption</u>			
	<u>In-Flux</u> mg/dm ²	<u>Out-of-Reactor</u> mg/dm ²	<u>In-Flux*</u> ppm mg/dm ²		<u>Out-of-Reactor**</u> ppm mg/dm ²	
Ti, Grade II	2.1					
	1.4	0.4	31	0.69	0	0
	0.7					
Nb	11.7					
	51.2	78	0	0	20	1.1
	60.0					
Nb-1% Zr	41.5					
	13.4	52	0	0	0	0
	79.9					
Zircaloy-2	11.1	11.0	27	0.5	42	0.78

*After 37 days in-flux.

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

Out-of-flux weight gains on niobium and niobium-1% zirconium were in general agreement with the highest weight gains on corresponding in-flux specimens but were high relative to Zircaloy-2 specimens in the same quadrant. In-flux and out-of-flux weight gains on titanium were approximately a factor of ten lower than Zircaloy-2 weight gains.

Titanium absorbed hydrogen in-flux but not out-of-flux. Hydrogen absorbed by the in-flux specimen exceeded the hydrogen produced by oxidation of the titanium. This appears to indicate permeation of the relatively thin oxide by radiolytic hydrogen. Niobium and niobium-1% zirconium did not absorb hydrogen in-flux. However, pure niobium absorbed 20 ppm out-of-flux after 200 days, at a weight gain of 288 mg/dm².

Specimens of titanium and niobium-1% zirconium and zirconium alloys are included in a 4-cycle, G-7 loop test to be discharged in March 1967.

Intercalibration Test of Counting Laboratories. The monitor materials for the intercalibration test have been irradiated in the MTR-VH-2 rabbit facility. The materials have been returned to Pacific Northwest Laboratory and are being counted prior to distribution to participating laboratories. The distribution has been scheduled for early February.

A two-dimensional transport-theory calculation of the MTR core and rabbit facilities has been initiated. Results of this calculation will provide spectral-averaged cross sections for the monitor materials. These average cross-sections will allow the participating laboratories to determine the fluence from the activated monitors.

Fracture Mechanics

Fracture testing and data analysis of irradiated A302-B pressure vessel steel DCB specimens was completed during the month. The estimated fast ($E > 1$ MeV) fluence was 4×10^{19} nvt which was attained in the ETR G-7 hot water loop at a temperature of 280 C. This fluence represents an upper limit to the expected lifetime exposures in the pressure vessel structure of proposed PWR and BWR designs.

The plane strain fracture toughness, K_{Ic} , of the irradiated A302-B was found to be considerably reduced over the temperature range of -196 C to 0°C. In general, the reduction of toughness from the unirradiated material amounted to 20-25 ksi $\sqrt{\text{in.}}$ over the entire test temperature range. These data represent the first observed effect of subtransition irradiation embrittlement in pressure vessel steels. For example, previous data from identical steel specimens irradiated to a fast fluence of 7×10^{18} nvt revealed no change in fracture toughness when compared with unirradiated A302-B data. There was, however, a decrease in the stress intensity at crack arrest in these specimens indicating that neutron irradiation to 7×10^{18} nvt only reduced the ability of the A302-B to arrest a running crack. Although the recent tests on the A302-B exposed to 4×10^{19} nvt reveal a further reduction in stress intensity at crack arrest, the stored driving energy for crack propagation in a structure irradiated to 4×10^{19} nvt would tend to be less than for lower exposures. Such a conclusion is based on the small difference observed between the fracture toughness (stress intensity at crack initiation) and the stress intensity at crack arrest after irradiation to 4×10^{19} nvt.

As mentioned in previous monthly reports, a set of experiments is in progress for the purpose of establishing size effects on fracture toughness and the limitations on sizes of DCB specimens for low-strength, high-toughness materials. A portion of these tests which are being used to determine the effect of side groove depth on the fracture toughness have been completed. These tests revealed that the measured or apparent value of fracture toughness is abnormally high if the side grooves are too deep or too shallow. The fracture toughness was found to be at a minimum and also considered to be valid, when the remaining or net thickness was from 25 to 40% of the gross thickness. The anomalously high values of fracture toughness can be attributed to excessive plastic deformation at the crack tip prior to crack propagation due to a lack of restraint provided by the side grooves. Qualitatively, the loss of restraint, resulting when the side grooves are either too deep or too shallow, is in agreement with a corresponding decrease in the stress concentration factor for the grooves. For example, elastic stress analysis reveals that the stress concentration effect in a doubly notched plate is maximum for a net

to gross width ratio of 0.3. In addition to providing some insight into the influence of side grooves in DCB specimens, these data will provide an invaluable aid in the design of future DCB specimens for testing pressure vessel steels.

Fast Reactor Supporting Studies

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

Two full EBR-II subassemblies containing candidate cladding and structural alloys have been completed. These subassemblies are to be shipped to the EBR-II site for charging in early February. One of the subassemblies, scheduled for 1×10^{23} fluence, is to be placed in a Row 2 position in the reactor core; the other will be placed near the inner blanket in Row 7 and is scheduled for 1×10^{22} fluence. Each subassembly contains a series of sodium-filled, gamma-heated subcapsules designed to operate at three temperatures; namely, 900, 1200, and 1400 F (480, 650, and 750 C). The capsules contain mostly 304 and 316 tensile, biaxial creep, and uniaxial creep specimens. Additional alloys, such as 348 and 321, are also included as well as Hastelloy X-280, Inconel 600, Incoloy 800, Inconel 718, Inconel 625, and Inconel X-750. Pure molybdenum, iron, and rhenium single crystals are included for basic damage studies.

Tensile tests are being conducted on annealed 304 stainless steel irradiated to 2×10^{22} fluence in the EBR-II in a helium environment. The objective of these tests is to establish temperature [RT to 1500 F (815 C)] dependence and strain rate dependence of tensile properties. The results will be compared to thermal reactor data to aid in establishing the thermal/fast reactor correlations needed to apply the large body of thermal reactor data to fast reactor technology.

Development of the biaxial sodium environment creep test apparatus for testing irradiated specimens and controls is continuing. A prototype of the stressing system, sodium filling, and sodium recovery system has been completed. This system has been successfully operated at room temperature using alcohol in place of sodium.

Test Facilities. The first phase of Building 321-A renovation is nearing completion. This will provide supporting laboratory facilities for the testing equipment. The second phase of the building renovation, which will include ventilation, will be done under a separate authorization for which a project proposal has been submitted.

The shielded prototype cell has been moved into place by the CPFF contractor. All lead brick is in place, and the structure is now ready for a radiation leak test.

The plans for the piping to serve the testing portion of the liquid metal (sodium) loop have been completed, and an information package is being prepared for offsite construction of this portion of the loop. The loop support facility, containing the E.M. pumps, heaters, and purification equipment, is now under construction by MSAR Corporation and is expected to be shipped in mid-February. The actual test units to be used in the liquid metal loop are now in the final design stages. This new type unit is simplified and much smaller (18" shorter) than the original prototype; yet the performance characteristics have been increased, especially in the precision alignment of the samples. Many of the improved parts used in the new test unit can be used in the test units to be installed in the shielded prototype cell. A device for the accurate measurement and safe dispensing of liquid sodium has been designed. Such a device is needed for measuring precise quantities of sodium and dispensing these quantities into test capsules of variable size and volume. The test capsules to be served will come from a variety of testing devices and will require accurate loading with widely varying volumes of sodium. Some parts of this device are now on order, and others are being machined.

A review of physical requirements, safety, and radiological requirements has indicated that it is possible to operate the shielded prototype cell with radioactive specimens in test before the building renovation is completed.

Liquid-Metal, Ex-Reactor Control Studies. The purpose of this phase of the program is to provide for the timely accomplishment of irradiation, testing, and subsequent processing of data obtained from a variety of reactor structural materials. Irradiations will be performed in aqueous, gaseous, and molten metal environments. Elevated temperature irradiations and tests will be emphasized. Special testing techniques applicable to either irradiated or unirradiated specimens will be developed.

EBR-II and ETR liquid-metal ex-reactor experiments and thermally-aged inert atmosphere control experiments are being prepared. These experiments will duplicate environmental (including liquid metal flow, where applicable) and thermal conditions of in-reactor irradiation capsules. Initial experiments are expected to become functional in mid-February 1967.

Liquid-metal and thermal-aging control capsule experiments are being initiated for the following experiments: GEH-22-01, an irradiation capsule containing 12 tensiles of each of annealed AISI 304 and 348 stainless steel, operating at 1200 F (650 C) with NaK environments; GEH-22-03, containing 16 tensile specimens of each of annealed AISI 304 and 316 stainless steel, operating at 1400 F (750 C) with sodium; and GEH-22-04, containing 16 specimens each of annealed AISI 304 and 316 stainless steel and 8 solution-treated AISI 348 stainless steel.

Thermal-aging control studies in the absence of sodium will be conducted for EBR-II irradiation experiments designated BNWL A1, A2, A5, and A6, which are being conducted in a helium environment.

Fast Neutron Mechanisms. Construction and preliminary testing of a heat transfer apparatus that will be used to study the effect of temperature gradients and thermal diffusion on interstitial solute redistribution and bubble motion in cladding alloys has been completed. With a few minor modifications, it will be possible to readily achieve the temperature gradients and mean specimen temperatures required for this work.

A series of room temperature hydrogen permeation and desorption experiments has been initiated using cold-worked steel specimens and electrolytic charging techniques as a precursor of hydrogen-defect interaction studies of irradiated alloys. Similar investigations are being conducted at intermediate temperatures (300-600 C) using gas phase charging methods to define surface effect behavior for subsequent cladding studies. Significant differences in the time constants for hydrogen absorption and desorption have been observed for specimens of typical cladding thicknesses. However, it has not yet been possible to satisfactorily determine the temperature and pressure dependence of the responsible surface effects, because of specimen temperature fluctuations associated with the nonequilibrium permeation technique used. A hydrogen supply system is, therefore, being developed that will use the unique pressure-temperature characteristics of hydrides to change the partial pressure of hydrogen in the permeation cell without affecting its heat transfer characteristics.

ATR Gas Loop Supporting Studies

Model Gas Loop Studies. The model gas loop is a 2100 F (1095 C) recirculating helium loop which is being operated to determine the effect of the helium atmosphere on materials and specimen holders, and to see if prolonged use will affect the operating characteristics of the new heater. This loop is a one-tenth size model of the gas loop that is being constructed in the Advanced Test Reactor at Idaho Falls.

The model gas loop has operated a total of 2197 hours since installation of the new heater with 47 thermal cycles from room temperature to various heater outlet temperatures. The horizontal test assembly has accumulated a total of 506 hours with 78 hours during the past month. The vertical test assembly was reinstalled in the loop, and pressure drop measurements were made at various temperatures and flows.

The loop was shut down most of the past month while the horizontal test specimens were removed for inspection. During this outage, faulty bearings in the French-made motor of the frequency changer were replaced, and the helium exhaust from the chromatograph and water monitors were connected into the storage balloon to conserve helium.

Specimens that are irradiated in the ATR gas loop will reach temperatures up to 2500 F (1370 C). A study is being made of methods to raise the specimen temperature within the vertical test section of the model gas loop in order to more closely simulate these temperatures. Three methods have been considered: (1) heating the gas with a resistance type heater, (2) induction heating the specimens, and (3) resistively heating tubular sections which would contain the test specimens. The third method has been selected as the one having the greatest potential of success and requires the least amount of development effort. This arrangement would utilize a number of molybdenum tubes that are electrically connected in series. The test specimens would be placed inside these tubular heating elements and completely insulated (electrically) from them. The cluster of tubes would be arranged to flow a portion of the helium over the specimens and bypass the remainder of the gas through the annular passage between the present test section and the auxiliary heater. The specimen temperatures will be controlled by varying the gas flow or the power to the auxiliary heater. A computer program will be used to determine the specimen temperatures that can be reached by this method under varying flows and power settings.

Thermal-Hydraulic Analysis. A steady-state heat transfer program was set up to calculate temperatures of a columbium hanger rod and test section liner under an assumed "worst case" condition in the ATR helium loop. Temperatures were calculated for a representative horizontal cross section through the test section. Thermal radiation effects were accounted for. Helium temperature was taken as 2000 F (1095 C), and a heating rate of 25 watts/g was assumed.

At the above conditions, the hanger rod temperature will be 2530 F (1390 C), the liner temperature will be 3450 F (1900 C), and the tube wall will be at approximately 950 F (510 C). Since the liner will not bear any structural load other than its own weight, the temperature, while it is high, should not cause any gross structural failure, because it is still well below the columbium melting temperature of 4475 F (2470 C). The strength of the columbium hanger rod at 2530 F (1390 C) will be investigated, and some means of lowering the rod temperature may have to be devised.

It is planned to run more computer cases to calculate hanger rod and liner temperatures with varying helium temperatures, materials, and heating rates.

Experimental measurements of the pressure drop across the test assembly in the model gas loop at several temperatures and gas flow rates have been obtained. Pressure drops at temperatures above 500 F (260 C) were higher than those obtained in similar experiments conducted two years ago. In both sets of experiments, however, the measured pressure drop multiplied by the helium gas density is a linear function of the square of the mass flow rate. This is the expected relationship for frictional losses in a fluid flow system. Extrapolation of the higher pressure drop data from the recent experiments to ATR operating conditions still leads to the conclusion that the maximum allowable pressure drop for the ATR specimen test assembly will not be exceeded with the current design.

Super Alloy and Refractory Metal Corrosion Studies. Corrosion coupons exposed in the ATR model loop (504-hr total exposure at temperature) continue to follow the weight change trend previously reported. Metallography and microprobe analysis of the samples taken from the high gas velocity section [2100 F (1150 C)] are presently being completed. cursory examination of the microstructure of Hastelloy X-280 and Haynes 25, total metal loss of 2.77 and 2.67 mg/cm², respectively, shows metal loss from grain boundaries and what appears to be subsurface voids. Niobium, 2.24 mg/cm² total weight gain, shows a continuous surface case approximately 8μ thick. The base metal also contains what appear to be intermetallic inclusions. After removal from the model loop, the tantalum sample, 1.77 mg/cm² weight gain, exhibited a loose, easily wipeable black deposit on the surface. However, metallography failed to show a well defined subsurface layer. The contaminant has yet to be determined for either niobium or tantalum.

Duplicate coupons were added to the test holder in place of those analyzed. The assembly will again be exposed to loop conditions.

Damage Mechanisms in Iron

The objective of this program is to determine how interactions between irradiation induced defects interact with moving dislocations to modify the mechanisms of plastic deformation in a metal. Radiation damage and the role of interstitial impurities in alpha iron are currently being investigated.

During this period, efforts have been devoted toward improving the reproducibility of resistivity measurements, report writing, and the completion of the first experiment on ultrasonic attenuation in a deformed sample.

The difficulty previously experienced in obtaining reproducible measurements of the resistivity of cold worked iron samples was traced to a faulty chopper in the electronic galvanometer circuit and an intermittent short in the wiring of the sample probe. The correction of these two difficulties eliminated the large fluctuations (5-10%) in resistivity, but further effort is being devoted to tracing the source of smaller fluctuations (approximately 0.1%) still present.

Time has been devoted to the preparation of several reports during this period. A topical report, "Evaluation of Solid State Purification of Molybdenum," by W. S. Bennett and F. A. Smidt has been completed and is in the final stages of publication. The first draft of a paper entitled "The Study of Thermally Activated Flow by Temperature Change Tests" by F. A. Smidt has been completed and is in the editing stage. It will be submitted to Acta Metallurgica.

The first ultrasonic attenuation experiment to study dislocation multiplication was completed during this period. An attenuation of ultrasonic pulses is known to be proportional to the product of dislocation density and the fourth power of loop length of mobile dislocations; thus, it is a sensitive measure of dislocation multiplication during the early stages of yielding. These first experiments were of a survey nature to check the operation of the

equipment and experimental procedure. The first experiment was performed on a Mo single crystal (Fe single crystals of the necessary size are not available yet), and the attenuation was found to rise slowly during elastic deformation and then to increase sharply in the pre-yield microstrain region. The crystal broke free from the epoxy bonding at a stress of 18 Kg/mm^2 and a strain of 0.065%, and the resulting shock wave shattered the transducer crystal, thus terminating the experiment. The equipment appeared to be working satisfactorily, but modifications in the crystal mounting technique must be made to study higher strains. Another experiment on a Ni single crystal was completed, but the data have not been analyzed yet.

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

ATR Gas Loop Support

Vendor Data Review. Vendor contract submittals through 4447 have been reviewed and comments submitted to the architect-engineer.

Analytical Instrumentation for the ATR Gas-Cooled Loop. Two systems are being assembled for which it will be necessary to be able to test their conformity with specifications. The first system is for the Transfer Facility. Of concern are the hygrometers, particularly the one measuring the output of the air drying system. This is specified to be capable of measuring to -150 F dewpoint. This is a very restrictive requirement, and it will be difficult to establish inspection procedures to determine that the instrument will meet our specifications. Equipment is currently being assembled by PNL to prepare gases of known moisture contents in the range of the lowest instrument range $0\text{--}1 \text{ ppm}$, which is equivalent to less than -105 F dewpoint. By passing moist gas through a cold trap at the proper temperature, the moisture can be reduced to any desired level. Such a trap and related equipment will be used for this testing. There are two additional hygrometers to be used on this facility that do not have to meet such restrictive specifications. These will be tested in a similar manner to establish that they meet specifications.

The second system, the analytical instrumentation system for the gas purification system, includes three hygrometers, two carbon dioxide analyzers, one carbon monoxide analyzer, one oxygen analyzer, and one gas chromatograph to measure hydrogen, oxygen, nitrogen, methane, and carbon monoxide. In addition to these instruments, there will be a total impurity monitor measuring the gas in the primary loop. Part of the system is a diffusion cell assembly to supply high purity helium as the reference gas for the above instruments. It also supplies pure gas to the dilution rotometers used for diluting the commercial standard gases to the low levels required to calibrate these instruments.

One of the major problems we have encountered with this system has been an incompatibility between the infrared analyzers for carbon monoxide and carbon dioxide recommended by Mines Safety Appliance for operation at 240 psi

and the diffusion cell assembly and rotometer package designed for operation at pressures of less than 100 psi. This presents a calibration problem since accurate calibration cannot be performed with this large a pressure difference. The solution of this problem is a vendor responsibility, but we have to be prepared to verify that their solution meets specifications. It is planned that we will do this by preparation of gases with the small concentrations of carbon monoxide and dioxide that the instruments are to measure and testing the instruments directly at their operating pressures. The oxygen analyzer has a built-in calibration system. The hygrometers in this system will be tested by the same methods described for the Transfer Facility.

One other area that needs some investigation involves the sample line components, specifically solenoid valves and "trace" heating of lines. The original specifications (T-3003) call for the use of Black Automatic Controls, Ltd., solenoid valves. These are a special flow-through type valve designed by the workers on the British Dragon project. They reported that all other valves they had tested were unsatisfactory. Several of the instruments being used in this system contain conventional skinner solenoid valves. The vendor has interpreted the specification on Black valves to extend only up to the instrument. However, it is the entire system that must operate according to specifications, and these valves may be limiting. To learn more about this problem, a test system has been designed and is being assembled. This will use a sensitive, rapid responding Panametrics hygrometer to measure rate of bleed down of valves and lines when a step change in moisture level is made. This will be done at various temperatures so "trace" heating can be evaluated.

The gas chromatograph and total impurity analyzer are undergoing test on our model gas loop. There are still a few problems to be ironed out, but these instruments appear to be capable of performing their specified functions.

Investigation of Type 304H Stainless Steel Longitudinal Pipe Welds. In compliance with a request by the Idaho Operations Office, gas tungsten-arc welds made by Swepeco Tube Corporation were evaluated by W. F. Brown, Battelle-Northwest. These welds were from a section of the pressure containment pipe for the ATR gas loop. The Solar Aircraft Company, who is fabricating the piping assembly, purchased the 12-inch, schedule 40, type 304H stainless steel piping from Swepeco Tube Corporation.

Swepeco's manufacturing process consists of forming plate stock into a tubular configuration and welding the longitudinal square groove butt joint using their gas tungsten-arc welding process called "Syncroweld." The Syncroweld process utilizes two torches positioned so that welding is performed from the underside of the joint as well as the upper side simultaneously without filler wire addition. This welding procedure yields a joint configuration comparable to a double-welded butt joint.

Evaluation of the 7-1/2-inch long weld was based on the results of visual examination, x-ray examination, fluorescent penetrant inspection, and metallographic examination. The criterion for evaluation was Section III, Nuclear Vessels, ASME Boiler and Pressure Vessel Code.

Fluorescent penetrant inspection revealed several surface cracks. Metallographic examination revealed that over 50% of the joint length had incomplete fusion penetration. Although diffusion bonding had occurred in the unfused areas from high temperature and pressure, the bond was of relatively poor quality compared with the fused area. Although x-ray could not detect this condition, the metallographic examination indicated that the process was out of control.

Based on this investigation, it was concluded that the welds submitted for evaluation were not acceptable and did not meet the intent of Section III of the ASME Boiler and Pressure Vessel Code.

A report (BNWL-CC-980) describing this investigation was prepared and distributed.

Visit to Fansteel Metallurgical Corporation. The Fansteel Metallurgical Corporation in Los Angeles, California, who is currently fabricating the ATR gas heater, recently found defects by radiography in the weld between the 12-inch nozzle and the heater shell. W. F. Brown, Battelle-Northwest, visited Fansteel to review their welding work and their repair procedures.

All of the welds with the exception of the 12-inch nozzle weld were free of detectable defects. Their repair techniques to date and their verbal description of repair procedures were satisfactory. Based on these discussions Battelle-Northwest has requested that they prepare and submit detailed welding and repair procedures for this joint.

Metallic Fuels Development (G. A. Last)

Three thorium - 2.5 wt% uranium (93.2% enriched) - 1.0 wt% zirconium tubular fuel elements clad in Zircaloy-2 are under irradiation in the P-7 loop in the ETR. The three fuel elements which were being irradiated in the M-3 loop have been replaced with another test and are being stored at the ETR site.

The current status of the test elements is summarized in the following table. The temperature data for fuel elements GEH-10-64, 65, and 71 are corrected for a temperature rise across an oxide buildup on the clad. The temperature data for the other fuel elements are uncorrected.

No.	Fuel Swelling %	Burnup		Maximum Core Temp. °C	Specific Power w/gm	Surface Heat Flux Btu/Hr-Ft ²
		Fissions/cm ³ MWD/T	At. %			
65*	2.9	5.7 x 10 ²⁰ (16,600)	1.90	410	38	5.5 x 10 ⁵
64*	1.8	4.0 x 10 ²⁰ (11,500)	1.32	455	30	4.3 x 10 ⁵
71*	1.0	2.5 x 10 ²⁰ (7,100)	0.81	490	44	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 (2,500)	0.29	440	33	4.8 x 10 ⁵
66**	1.6	4.1 x 10 ²⁰ (11,850)	1.36	445	36	5.4 x 10 ⁵

* Heat generation conditions during ETR Cycle 85.

† Heat generation conditions during ETR Cycle 84.

** Returned to Battelle-Northwest for examination. Heat generation conditions during ETR Cycle 77.

High Exposure Uranium Irradiation Test

Irradiation of 24 hollow core uranium fuel elements in the M-3 hot water loop of the ETR continued during reactor cycle 86. Calculated exposure on the maximum power elements is approximately 2000 MWd/ton at the end of cycle 86. Calculated maximum fuel core temperatures range from 390 C in the lower power elements to 545 C in the maximum power element. These fuel elements are 0.45 inch diameter by 6.25 inches long and are clad with either 0.025 inch or 0.050 inch Zr-2. 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% is being used to achieve the desired burnup rate (10,000 MWd/ton in one calendar year) and temperatures.

Fabrication and preirradiation measurements were completed on four additional fuel elements which have been shipped to the ETR to replace certain selected elements. Efforts are being made to arrange to ship several elements discharged at the end of cycle 86 to the Battelle Columbus Research Reactor for neutron radiography, in order to provide some insight into what is occurring in the uranium around the control voids, without having to destructively examine the fuel element. Following the radiographic tests, the elements will be returned to the ETR for additional irradiation.

Brazing was completed on nine hollow core fuel elements being prepared as replacement elements for those now being irradiated. The brazing was accomplished with no rejects, and radiographs taken after brazing showed uniformly bonded joints with no porosity in the braze. After the elements were electron beam welded, however, the radiographs showed extensive porosity in and adjacent to the weld closure, although the welds appeared good on the surface. The weld areas were machined back to expose the voids, and the rods were rewelded using a modified procedure designed to prevent excessive heating of the braze joint. The radiographs, however, again showed porosity in the weld areas.

No explanation has yet been found for the sudden appearance of the weld porosity problem. In future brazes the braze will be held molten in vacuum for a longer period to allow more time for outgassing, and welding will be done by tungsten-inert gas welding rather than vacuum electron beam welding in an effort to eliminate this problem.

Fast Reactor Dosimetry and Damage Analyses (R. E. Nightingale)

EBR-II Dosimetry Experiment

Four experimental capsules containing a variety of neutron dosimeters were irradiated in a recent EBR-II experiment. The objectives of this experiment were to aid in evaluating cross sections for monitors which could be useful in materials irradiations and to measure flux gradients and spectral shifts within irradiation facilities. These experiments, conducted during a

low power run, provided an opportunity to use a number of monitors that could not be used during routine testing because of short half-lives of the reaction products, and to experiment in positions within the reactor that would not be available otherwise.

This experiment was conducted on January 17th during low-power, short-term, EBR-II physics test runs. The reactor operated for 30 kW-hr. Monitors were irradiated in pins within special test subassemblies located in the center, Row 3, and Row 6. Three of the four capsules contained seven, 14-inch long, 0.020-inch diameter wires. The monitors in three capsules were iron, nickel, titanium, aluminum, aluminum-1% cobalt, aluminum-1% silver, and copper wires. The fourth capsule was located in the center subassembly and contained depleted uranium, cobalt, natural uranium, scandium, and zinc. The following table lists the reactions, product half-lives, and sensitivity energy ranges for each material included in this equipment.

<u>Monitor Material</u>	<u>Reaction</u>	<u>Product Half Life</u>	<u>Energy Range, MeV</u>
Ni	Ni ⁵⁸ (n,p)Co ⁵⁸	71 d.	>4.1
Fe	Fe ⁵⁴ (n,p)Mn ⁵⁴	313 d.	>4.3
Ti	Ti ⁴⁶ (n,p)Sc ⁴⁶	84 d.	>6.0
Cu	Cu ⁶³ (n,α)Co ⁶⁰	5.24 yr.	>9.2
Al	Al ²⁷ (n,α)Na ²⁴	15 hr.	>7.5
Zn	Zn ⁶⁴ (n,γ)Zn ⁶⁵	245 d.	0.003
Sc	Sc ⁴⁵ (n,γ)Sc ⁴⁶	84 d.	0.004
U-238	U ²³⁸ (n,f)	4.5x10 ⁹ hr.	1.4
Al-1% Co	Co ⁵⁹ (n,γ)Co ⁶⁰	5.28 yr.	10-2
Al-1% Co	Ag ¹⁰⁹ (n,γ)Ag ^{110m}	253 d.	10-5

A similar experiment will be conducted in February. The results of these two low-power, short-run experiments will aid in the selection of suitable materials and interpretation of data in full-power, full-cycle irradiations.

High Temperature Fluence Monitors

The study of the effects of neutron irradiation on materials and fuels, and the use of this information for reactor design require that flux be accurately measured. Sometimes the flux must be measured in locations where the monitor cannot be cooled, and monitor temperatures may exceed 1200 C. The number of metallic monitors that will survive this temperature and not react with their environment is limited.

The use of ceramic pellets composed of metallic oxides in an aluminum-oxide diluent has been reported. However, the neutron-sensitive metal concentration varied from sample to sample. This variability requires that each

individual monitor pellet be precalibrated by neutron activation before it can be used for a test. Calibration proves time consuming and impractical where hundreds of monitors are needed.

An attempt to develop high temperature ceramic monitors that are both metallurgically stable and contain a constant quantity of metal has been started. Initially, an attempt will be made to develop a metallic oxide - magnesium oxide that will remain stable up to 2400 C. The first pellet, containing iron has been fabricated, sectioned, and placed in a reactor for activation. Measurement of the activity of each section and metallographic studies will determine if an even distribution of metal has been obtained. Development of the fabrication process is continuing, and several hundred pellets containing iron, nickel, and cobalt will be produced and tested.

ENGINEERING DEVELOPMENT

Neutron Flux Monitors (W. G. Spear)

Regenerating Detectors (Thermal Flux)

Test program authorizations and capsule design information are being obtained for the ETR in-core testing of a set of regenerative thermal neutron flux detectors. ID-27 application forms requesting test program authorization were prepared, and arrangements were made at the ETR to perform measurements establishing the capsule lead length. The detector cable length will be determined by inserting a physically similar dummy capsule into the reactor core.

Blending and final preparation of an additional 90 milligrams of U-234 into the sensitive regenerating coating used in another set of two detectors is progressing. During operation the U-234 isotope transmutes by thermal neutron capture to U-235 thereby replacing the burnup loss. Because of this regenerative action, nearly constant detector sensitivity is achieved for in-core use.

In experiments, these detectors have demonstrated relatively constant sensitivity during prolonged periods of irradiation at a particle fluence of about 7×10^{21} . An interim technical report concerning the regenerative detector operating performance at high neutron flux levels is being prepared.

Beta Current Generator Detectors (Thermal Flux)

The development and extensive testing of B-11 self-powered neutron detectors is the subject of an interim technical report being prepared. The described detector features a balanced twin chamber, twin lead design for achieving gamma compensation. In an operating reactor the B-11 sensitive

material reacts with neutrons (n, γ) to form the short-lived B-12 isotope which decays rapidly with the emission of energetic beta particles. These emitted beta particles constitute a measurable electric signal related in magnitude to the incident neutron flux level. The small output signal observed during recent experiments with this detector was related to the new, lower (by a factor of 10) cross section of 0.005 barn for this reaction and recently reported at another laboratory. An analysis of all experimental data and theoretical parameters is being conducted to completely define the design requirements and operational characteristics of the beta current detectors.

Beta Current Generator Detectors (Fast Neutron Flux)

The Be-9 isotope is being investigated as a candidate material for fast flux detection. When subjected to incident fast neutron flux, this isotope undergoes an (n, α) reaction with about 0.1 barn cross section at 3 MeV and providing a beta particle emission (~ 3.5 MeV) with a relatively short half-life of 0.8 second. This beta emission constitutes a signal current proportional to the neutron flux level. In an experimental chamber, beryllium in an oxide or powdered metal state would 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 high gamma levels of 10^{10} R/hr, the balanced twin lead twin chamber concept developed in the thermal flux detector program will be extended.

Evolved Gas Detection Concept (Fast Neutron Flux)

In this unique concept of fast neutron detection, an evolved hydrogen or helium gas would be collected from a very simple capsule and evacuation tube immersed in the reactor core. Gamma interference problems would be minimized by the location of electronic and other instrumentation in the less destructive environment external to the core. For this detector, (n, α) or (n, p) reactions with B-10 or other materials would release helium or hydrogen in quantities proportional to the neutron flux. As an estimate, about 10^{11} atoms/sec of helium gas evolves from one gram of B-10 at an incident flux level of 10^{13} nv. For this quantity, the evolved gas will be measured by a mass spectrographic or optical spectrometer type instrument. It is expected that a flux of 10^{10} nv or greater could be detected using these evolved gas concepts.

Microwave Detectors (Thermal Neutron Flux)

The resonant frequency of microwave cavity containing a plasma is a function of the plasma-free electron density. If the plasma is created within the cavity by an incident neutron flux, an equilibrium electron density will be established, its value determined by electron generation rate and the competing dissipation mechanisms. Using this method, the neutron flux density can be measured by monitoring the cavity resonant frequency shift. The following methods can be used to produce the required plasma: (1) For thermal flux, a

gas such as He-3 can be used where an (n,p) reaction with subsequent ionization of the gas by the charged particles creates the plasma, and (2) a material such as U-235 or a special regenerating mixture of isotopes could be used where the fission fragments cause gas ionization.

To avoid loss of sensitivity experienced when a dielectric capsule (quartz) was used to enclose the detector gas, a gas-tight seal to the microwave cavity is being sought. Eight vacuum tight waveguide sections are presently available, three of which will be brazed to fabricated cavities. These three cavities, each of slightly different design, are presently being modified slightly prior to their preparation for brazing in the recently completed dry hydrogen atmosphere brazing facility. If successful, the assembled cavities will be positioned within tubing required for final in-reactor experiments of the thermal flux detectors. Initial attempts to obtain satisfactory brazes in a hydrogen atmosphere were partially successful, and with slightly improved techniques vacuum joints are anticipated.

One of the three cavities will be used to obtain a non-dielectric resonant enclosure by using metalized discs brazed directly to the cavity coupling apertures. This effort is in parallel with the use of sealed waveguide sections mentioned above.

Microwave Detectors (Fast Neutron Flux)

Microwave techniques used to measure fast flux density will be similar to methods proven successful for the measurement of thermal neutron flux. A measurable shift of the resonant frequency of a microwave cavity would be caused by ionization of a cavity-contained gas. It is expected that the gas would be ionized by reaction fast fission fragments of an appropriate isotope, such as high purity U-238, placed within the cavity. Inasmuch as cross sections are small and gamma high in such a reactor, compensation for the gamma interference must be achieved. This can be done by locking the microwave oscillator frequency to a gamma reference cavity. Any difference frequency between the reference and neutron-sensitive cavities would then be related to the fast neutron flux.

An additional method of measuring fast flux would be to relate the incident flux and a gas-induced frequency shift of a microwave cavity through which gas from a beryllium fast flux reaction is passed. The amount of gas evolved would depend upon the flux density and would be related to the magnitude of the cavity frequency shift resulting from gas molecules within the cavity. Theoretical considerations indicate an instrument sensitivity of about 10^{11} nv for a reasonable gas evolution rate of 10^{-2} atoms/sec/nv.

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

Microwave Detection of Impurities in Coolant Gases

The moisture content of an otherwise pure gas will change the dielectric constant of the gas. Thus, the presence and amount of water vapor can be determined by measuring the frequency shift of a microwave cavity or by measuring the phase change in a section of waveguide through which the gas flows. In the latter case, it may be possible to use the process gas piping as the waveguide, thus becoming a truly in-line device. Although measurements at room temperature indicate a sensitivity of about 900 Hz per ppm at the lower water vapor concentrations, this may be due to difficulty in accurately reading the dew pointer used to measure water vapor content. The value of 75 Hz per ppm obtained for high concentrations of water vapor is more reliable since theoretically, the frequency shifts should be linear and about 125 Hz per ppm. The dielectric constant of water vapor is a function of temperature as expressed in the following equation:

$$V \frac{k-1}{k+1} = A + \frac{B}{T}.$$

At room temperature, the second term on the right dominates, and the dielectric constant varies inversely with temperature. At temperatures of 600 C, the second term is still about five times as large as the first term, and the dielectric constant is still temperature dependent. Thus, any measuring system must be compensated for temperature variations. A detection system for detecting the presence of moisture will still be effective but will lose some sensitivity.

Future efforts will be directed toward use of process piping as waveguide and to the measurement of high temperature gases. Also, a long term stability test will be conducted on the present measuring system.

High Temperature Measurements

Measurement of temperature within high temperature reactors expected to be operating in the future poses severe environmental problems with the sensors and associated interconnecting leads. A method of measuring high temperatures using microwaves is being investigated for its sensitivity, linearity of response, and potential for long operating lifetime.

A completed microwave assembly, including the input waveguide and resonant cavity, were successfully tested during initial experiments regarding the shift-of-frequency method of monitoring temperature. After evacuating the assembly, the microwave cavity was immersed in an oven and its resonant frequency monitored as a function of its temperature. Linearity of response was excellent over the temperature range 25 C to 680 C with a sensitivity of 290 kHz per degree centigrade. A nearly straight line response was realized with minute variations probably due to inaccuracy in reading the wavemeter.

A second temperature monitor assembly received from an offsite vendor should allow measurements to 1000 C to be performed in the near future.

Infrared Techniques

The electronic control unit for the second hygrometer (Mark II) has been fabricated and tested. This unit provides master control over all functions of the hygrometer and contains circuits to amplify the error signal and to control the motors and servo system.

Fabrication work on the main hygrometer unit was resumed so that it will be completed and ready for test and evaluation in February 1967.

Effort has been expended in the past month in the exploration of a different hygrometric design concept. In this concept the hygrometer consists of a light source, a 12-aperture chopper wheel, a reference and a sample chamber, a lead sulphide detector, a frequency selective difference amplifier and indicators, and necessary optical elements. Holes in the chopper wheel which revolves at 30 rps are so spaced that energy from the source alternately passes through the reference and sample chambers at a rate of 360 Hz. The detector is energized by light traveling through the chambers. With no difference in chamber moisture content, the output of the detector is zero. When the sample chamber contains moisture, the difference signal is amplified and displayed.

Upstream Boiling Burnout (D. R. Dickinson)

A new series of tests to study upstream boiling burnout and related phenomena was started and is now in progress. The principal objective of this present series of tests is to study the flow pattern transition observed earlier to precede the occurrence of upstream boiling burnout. The test section consisted of a 0.435-inch ID tube heated uniformly over a 6-ft length. Cooling water at 1500 psig was passed through the tube. In one series of runs with coolant mass velocity of 3×10^6 lb/hr-ft² and inlet qualities up to 25%, sharp drops in the surface temperature (5-8 F at heat fluxes of 200,000 to 300,000 Btu/hr-ft²) occurred on gradually increasing power at heat fluxes as much as 20% below burnout. These temperature drops occurred first at the downstream end, and then in some cases were noticed part way upstream at subsequent power increases. This evidence of a transition in flow regime supports previous evidence from reduction in pressure drop. This flow pattern change is believed to be a possible cause of the upstream burnout phenomenon.

PLUTONIUM UTILIZATION PROGRAM (F. G. Dawson)Fuels DevelopmentUnderwater Equipment

Underwater equipment items for the examination of FERTF fuels have been procured and/or fabricated. An underwater positioner and fuel storage racks have been designed and fabricated onsite. A scope for underwater visual inspection was received onsite. Procurement of all other items is under way. Beneficial use of the new equipment, after installation in the PRTR basin, is expected by March 1, 1967.

FERTF Testing

Critical tests were performed in the PRTR using a six-rod FERTF test element in rings 4 and 5 and a mocked-up eight-rod element in the central channel (1550) of PRTR. Results from these tests indicate that the best power match between the FERTF test element and the Batch Core elements in ring 1 can be achieved using an eight-rod cluster design for the FERTF test element.

Results of the critical tests, analyzed by members of the Light Moderator Reactor Physics Unit, were used to design an eight-rod FERTF test element that will provide the desired operating conditions. The design incorporates several features which permit minor adjustments in power generation in order to optimize the power match between the FERTF test element and elements in ring 1 of the Batch Core. Two FERTF test elements are being fabricated--one for thermal hydraulics studies and wear-fretting tests and one for critical tests in PRTR to verify the rod powers calculated for this design.

PRTR Fuel Element Cleanup

Because of the presence of foreign material in the primary cooling system of the PRTR, it was decided to completely discharge the reactor, flush and filter the primary cooling system, and clean, inspect, and re-band all of the elements before starting the Batch Core Experiment. The campaign to clean a total of 70 elements was started on swing shift, January 9, and was completed in seven shifts (4 swing and 3 days).

Debris from each element was collected in separate sample vials, and comments about each element were recorded on individual data sheets. With the exception of the replacement of one rod in one element, all of the elements appeared to be in excellent condition.

Most of the band damage observed appeared to be caused by the fuel element catching on a flanged joint during charging. To eliminate this damage, a "charging" funnel was designed and fabricated to guide the fuel element past the flanged joint. This funnel was used successfully during recharging operations.

Instrumented PRTR Fuel Elements

Debugging and final checkout of fuel rod pressure readout equipment as a complete unit indicated the need for a simpler and more reliable setup. The electrical complexity of the system and the lack of readily available repair components made the system difficult to troubleshoot and repair. A simplified instrumentation setup was designed and is being fabricated for immediate installation.

High Exposure Plutonium Studies

Gamma and x-ray energy spectra analyses were made on three plutonium samples recovered from Yankee reactor fuel. The samples selected were from a high ($\sim 20,000$ MWd/Mt) burnup, asymptotic neutron spectra fuel, a medium burnup ($\sim 11,000$ MWd/Mt) asymptotic spectra fuel, and a high burnup intermediate spectra fuel. The purified plutonium product solutions show distinctly different gamma energy spectra, particularly in the amounts of Am-241 and Pu-238 present.

The gamma spectra indicate the amount of Am-241 is 100% greater in the plutonium from the medium burnup asymptotic spectra fuel than from the high burnup intermediate spectra fuel and 50% greater than from the high burnup asymptotic spectra fuel. A part of this variation is due to length of time between plutonium separation and gamma energy analysis. The data will be verified by mass spectrometer analysis and by additional gamma energy analysis at specified dates to verify the Am-241 growth curve.

The plutonium from the high burnup intermediate spectra fuel appears to have the greatest amount of Pu-238, about twice that of the other two samples. More complete analysis of this data will be made with the help of a computer program and mass spectrometer analyses.

One kilogram samples of Pu metal, PuO₂, and PuF₄ were received for neutron dosimetry measurements. Also, one, ~ 50 -gram sample of PuO₂ from Shippingport fuel was received for dosimetry studies. The Pu-240 content of all these samples exceeds 20%.

The Phoenix PRCF fuel plate program was completed, and three Pu-Al reject plates are available and will be used for neutron dosimetry studies.

All analytical data are complete on the Shippingport fuel with the exception of the Am-243 and Nd-148 both of which are being carried on as development programs to assure the best analytical techniques will be used.

Fabrication Cost Studies

Detailed cost studies were developed recently at Battelle-Northwest laboratories for pellet filled or vibratory compacted fuel rods composed of natural uranium fuel material, enriched with either uranium-235, low exposure plutonium, or high exposure plutonium. Six different operating conditions were studied. Further studies have recently been made to determine the influence of plant throughput on fuel cost. Unit costs for 1/4- and 1/2-ton of fuel/day capacity plants were estimated for vibratory compacted UO_2 , enriched with either U-235 or high exposure PuO_2 . Additional cost analysis is under way to determine what cost savings might result from delaying the introduction of PuO_2 into the fabrication process for as long as possible.

Reactor Physics

D₂O Moderated Systems

Photoneutron Effects. Calculations of photoneutron production in D_2O moderated reactor systems have continued. Previous calculations were made assuming a spatially flat gamma source and assuming the space energy distribution of gamma rays is independent of the moderator and coolant temperatures.

A gamma source that varies spatially (according to the neutron fission density) was used in these calculations. The magnitude of the photoneutron production differed by about 5% from that which was obtained when a flat source was used. By including the effect of the moderator and coolant temperatures on the gamma ray distribution, the results changed by ~5%.

Analytical Correlation of PuAl Burnup Data. Calculations have been made for a PRTR cell containing a plutonium-aluminum 19-rod cluster irradiated to various exposures. The isotropic compositions determined from experimental analysis are used in the calculations, and the neutron spectrum is computed using HRG and THERMOS. Integrals used in calculating effective cross section ratios are obtained from these calculations. The effective cross section ratios are compared to values deduced from experimental burnup data. The calculations are based upon the use of various evaluated cross section sets. The purpose of this study is to determine which cross section sets coupled with the theory, agree with the experimental ratios.

Batch Core Experiment in the PRTR. Analysis of Batch Core critical tests is continuing. The data from the final axial flux traverse, using gold foils, have been analyzed. A new analysis is being made of lutetium foil data from the Batch Core critical tests in both the PRTR and the PRCF, to correct errors which were discovered in the original analysis.

Additional critical tests have been conducted with the Fuel Element Rupture Test Facility (FERTF) element in the PRTR. Gamma scanning of irradiated fuel rods from the FERTF element and from an element in the first ring of the Batch Core has been done to determine relative fission rates. Two of these tests were run with a six-rod cluster of $\text{UO}_2 - 2 \text{ wt\% PuO}_2$ in the FERTF, in channels 1849 and 1847. Other tests were run with an eight-rod cluster of $\text{UO}_2 - 2 \text{ wt\% PuO}_2$ in the FERTF, in channel 1550. A stainless steel basket tube having a wall thickness of 1/8 inch was used in all these tests. The eight-rod cluster was measured with both aluminum and stainless steel plugs in the center. The results of these tests were used to normalize calculations made to optimize the power match between the FERTF element and the rest of the Batch Core. Additional critical tests have been scheduled with the eight-rod assembly.

Calculations have been made to estimate the power match between a 19-rod fuel element in channel 1550 and the elements in the first ring of channels in the Batch Core in PRTR. The study included $\text{UO}_2 - 2 \text{ wt\% PuO}_2$ rods, in both H_2O and D_2O coolant, and $\text{UO}_2 - 1 \text{ wt\% PuO}_2$ rods in D_2O coolant, in channel 1550. The conclusions were that an H_2O cooled $\text{UO}_2 - 2 \text{ wt\% PuO}_2$ 19-rod cluster in channel 1550 would operate at about 8-11% higher power than the elements in the first ring.

Five samples of borated D_2O have been delivered to the Analytical Laboratories of the Chemistry Department for the determination of boron content. These measurements will serve to test the capability of measuring small concentrations of boron in D_2O and to compare an analytical determination with the TTR reactivity determinations.

Gamma Scanning Facility. A collimator modification has been completed, and the resolution of several NaI (Tl) crystals has been checked in preparation to gamma scan two cermet fuel pins which were irradiated in EBR-II. A 1/16 inch diameter aperture has been installed at the lower end of the collimator to improve the spatial resolution of the scanner. In checking the detecting system, it was learned that the resolution has deteriorated. The resolution of a number of NaI (Tl) crystals are being measured to determine a replacement. Plans are also under way to obtain a Ge(Li) detection system for the facility. The increased resolution of the Ge(Li) system will allow more detailed burnup information to be obtained nondestructively.

Effective Cross Sections for PuAl Fuel. The analysis of plutonium data now in progress requires an estimate of the ratio of Pu-241 decay to Pu-241 loss by neutron absorption during the period of irradiation. That is, the ratio $\lambda^{41}/\hat{\sigma}_a^{41}\bar{\phi}$ must be determined where $\lambda^{41} = 1.66 \times 10^{-9} \text{ sec}^{-1}$ is the Pu-241 decay constant, $\hat{\sigma}_a^{41}$ is the flux averaged cross section, and $\bar{\phi}$ is the average neutron flux during the irradiation. This decay term varies widely for samples cut from an element since the average flux, $\bar{\phi}$, changes rapidly as both a function of position and exposure of the sample. Values of $\lambda^{41}/\hat{\sigma}_a^{41}\bar{\phi}$ varied from a low of 0.02 to a high of 0.10 for the 1.8 wt% plutonium in aluminum samples irradiated in the PRTR, and $\lambda^{41}/\hat{\sigma}_a^{41}\bar{\phi} = 0.05 \pm 0.025$ seems to be a reasonable

average for the complete set of these samples. To obtain these values, $\bar{\phi}$ was calculated for each element from heat flow data by the formula

$H(\text{MWd}) = kV\Sigma_f \bar{\phi} \Delta t$ where $k = \text{MWd/fission}$ and Δt is the time that the element was irradiated. The macroscopic fission cross section, Σ_f and $\hat{\sigma}_{a41}$ were obtained by calculational methods for a simulated PRTR 1.8 wt% PuAl cell of volume V .

H₂O Moderated Systems

Analysis of PuO₂-UO₂-H₂O Lattices. Calculations have been performed to provide analytical correlations of data from recent PRCF experiments with UO₂ - 2 wt% PuO₂ fuel rods and H₂O moderator. The change in k_{eff} produced by voiding the moderator has been calculated for two loadings: one loading of rods whose plutonium contains 8 wt% Pu-240, and another of rods whose Pu contains 24 wt% Pu-240. The water-to-oxide volume ratio was 1.83 for both cores. Cell average cross sections, calculated with the codes THERMOS, HRG, and TEMPEST, were used in a 4-group diffusion theory calculation using HFN. Calculations were performed for voids of 5, 10, 15, and 30%, defining the voids as uniform reductions of H₂O density. A calculation was performed in which a discrete void region (15% of moderator volume) was included in the THERMOS geometry, to represent the experimental geometry more exactly. No appreciable difference was observed between the worth in this case and the case in which a uniform density reduction was assumed.

The measured void worths do not take into account interaction between cells. To determine how large this interaction effect is, the worth of a unit cell containing a 15% moderator void was calculated as a function of core radius. It was found that the sum of the worths of individual voided cells is ~30% smaller than the worth of simultaneously voiding all cells. When this interaction effect is taken into account, measured and calculated worths of moderator voiding are in good agreement.

Changes in k_{eff} due to changes in the moderator height are being calculated. Preliminary calculations overestimate the worth of moderator height changes by ~25%.

Cosine curves have been fit to calculated (HFN) axial flux and power distributions. The resulting axial bucklings are in good agreement with bucklings determined from measured distributions of activation and power.

Analytical Correlations of the Saxton Critical Experiments. The calculation of results to be compared to results from the Saxton critical experiments conducted at the Westinghouse Reactor Evaluation Center has been essentially completed. Both one and two zone fuel loadings have been included in the study. Values of the effective multiplication factor, k_{eff} , have been calculated using the THERMOS, TEMPEST, HRG, and HFN series of computer codes. The maximum difference of 4.5% between experimental and calculated values of k_{eff} , reported

last month, (the case for UO_2 fuel in a 0.792" lattice) was due to an error in the U-238 density. Correcting this error reduced the calculated k_{eff} by 2%. The calculated values of k_{eff} differ from experiment by at most $\pm 1/2\%$.

The effect of using a one dimensional diffusion theory code (HFN-cylindrical geometry) to analyze square loading criticals has been studied. As expected, the calculated values of k_{eff} are somewhat higher than a similar analysis of cylindrical loadings. In both cases the fuel volume was conserved in the resulting cylinders. Using a two dimensional diffusion theory code, EXTERMINATOR, in x-y geometry to analyze two square loadings, the calculated value of k_{eff} was comparable to the value obtained using HFN.

Two cores each containing zones of UO_2 and $\text{PuO}_2\text{-UO}_2$ fuel were analyzed using HFN. In each case the volume of a square fuel region was conserved, i.e., the concentric cylinders have the same volumes as in the square loadings. The calculated k_{eff} differs from experiment by less than 1.0% in most cases.

Reactivity Effect of PuO_2 Particle Sizes

The changes in reactivity due to differences in the size of the PuO_2 particles in mixed oxide reactor fuels ($\text{PuO}_2\text{-UO}_2$) are difficult to predict and can be important in the over-all evaluation of reactor loading. A series of experiments has been started to measure, experimentally, the effect that the PuO_2 particle size has on reactivity. The experiments will determine the change in reactivity for a PuO_2 particle size variation from zero to 700 microns for fuel containing about 24 wt% Pu-240. The oxide composition of the fuel is 2.0 wt% PuO_2 .

Four fuel rods containing 350-micron PuO_2 particles in UO_2 pellets and five rods containing $\text{PuO}_2\text{-UO}_2$ solid solution pellets were completed. The four UO_2 - 2 wt% PuO_2 fuel rods contained 350-micron particles which were prepared by jet-milling high energy impacted PuO_2 . The PuO_2 in the two sets has an isotopic content of 8 wt% $^{240}\text{PuO}_2$.

Since the effect of the particle size is mostly a fuel rod effect and somewhat independent of the moderator, the first experiments are being made in graphite lattices with simple geometries. The accuracy of the results is expected to be better than that obtained with an H_2O moderator due to the ease in handling the solid moderator and the reduced sensitivity to moderator displacement. A later comparison with water moderated lattices is to be made. All experiments will be conducted in the Physical Constants Test Reactor (PCTR).

The first experiment was in a 4.0-inch square graphite lattice. The fuel rods available for this experiment contained about 8 wt% Pu-240 and particle sizes of 0, 200, and 350 microns. The analysis of this experiment is incomplete, but preliminary results show a decrease in reactivity for an increase in particle size from 0 to 350 microns. The decrease is compatible with a calculated decrease of 0.028 in the neutron multiplication factor.

A work order has been placed for the fabrication of the water tank to be used in the core of the PCTR for measurements on light water moderated lattices.

PCTR Water Tank Calculations

A series of simple one dimensional diffusion theory calculations have been completed for unpoisoned light water moderated mixed oxide lattices loaded in the inner tank of the proposed PCTR light water test assembly. A range of hexagonal pitches from 0.66 to 1.32 inches were investigated for these 2 wt% PuO₂ (8% Pu-240) - UO₂, 1/2 inch in diameter, mixed oxide rods. These calculations indicate that the pitch at which the k_{eff} of the PCTR is a maximum can be modified from about 0.9 to 1.0 inch (as compared to 1.15-inch pitch fully water reflected) by using various combinations of PCTR drivers and natural uranium buffers in the graphite immediately surrounding the test cavity. Also, the fast-to-thermal flux ratio was found to be constant over, at least, the central 14 cm of the inner tank for all cores and pitches. This indicates that the spectrum in this portion of the test lattice is indeed representative of the mixed oxide cell. Some small difficulty has been encountered in correctly predicting PRTR driver and control rods worths. This will have to be cleared up before proceeding to optimize a driver and buffer combination for the proposed 1.0 inch square experiment.

Code Development

Maximum Likelihood Nonlinear Correlated Fields (Battelle-Northwest Program Likely). Two man-years of theoretical physics work on generalization of least-squares fitting logic, to meet the needs of the Plutonium Recycle Program, progressed to successful completion during the past month. Comprehensive documentation of the theory improvement has gone to press.

Offsite Distribution of Computer Programs. In response to a request from the Oak Ridge Operations Office of the Atomic Energy Commission, the latest active version of Battelle-Northwest program Learn (Maximum Likelihood Nonlinear Fields) was shipped to Oak Ridge for exploratory application to optimization of research and development plans.

In response to a request from the University of Washington, the latest active version of Battelle-Northwest program Likely (Maximum Likelihood Nonlinear Correlated Fields) was transmitted to their Seattle campus for student use in their graduate physics laboratories.

Synthesis Methods for Burnup Analysis. In response to USAEC invitation two Battelle representatives from Theoretical Physics participated in a three-day conference on Synthesis Methods for Burnup Analysis, sponsored by the Argonne National Laboratory. On a national planning basis spanning foreseeable

computer capabilities, the conference reached a majority agreement that the domain of reliability for synthesis methods is limited to criticality predictions, that synthesis schemes cannot be solely relied upon for power distribution and transmutation design, and that development effort must continue on improvement of brute force two-dimensional and three-dimensional methods.

Program CHAD. Atomic International's computer code CHAD (Code to Handle Angular Data) has been imported and has been made available for general use on the IBM-7090 computer. CHAD requires version 13 of IBM 7090 Fortran IV.

CHAD produces legendre scattering coefficients from angular data into legendre scattering coefficients in either the laboratory or the center-of-mass frame of reference. It also calculates the average cosine of the scattering angle in the laboratory system and the average logarithmic energy decrement per elastic collision.

Program CHECKER. The computer program CHECKER was imported from Brookhaven National Laboratory for use in checking input destined for inclusion in the ENDF/B evaluated neutron cross section library.

Improvements on HRG (Hanford Revised GAM). An auxiliary code to write an HRG data tape in decimal card image form was written and put in operation. By creating a decimal card form of the data tape, this code facilitates the conversion of HRG, particularly to computers with tape incompatibility with the IBM 7090 or Univac 1107. (Writing this code was prompted by difficulties arising in such a situation.) The input provides positive identification of the cards and furnished sufficiently detailed information of format and card-and-word count to enable a user to reconstruct a binary tape readily.

Program UNICORN. The computer program UNICORN has been modified to allow the use of a different energy mesh in the calculation of neutron cross sections using Briet-Wigner formalism. The new mesh allows the user to obtain a greater number of energy points in the valley between resonances than was previously possible when the maximum number of points to cover the peak of each resonance as specified. The new scheme inserts 45 additional energy points into each decade using an equal increment of 0.2, starting at 1.0 and ending at 10.0. The additional points help mainly at low energies where increased coverage of isotopes like U-238, with a large mean level spacing, is required.

The new scheme of calculating the energy mesh completely eliminates the requirement of supplying resonances with a zero neutron width in order to obtain additional energy points at desired energies.

Precise Evaluation of Differences of Error Functions. The calculation of thermal neutron scattering cross sections from simple theoretical models often leads to expressions involving the difference of two error functions multiplied by an exponential. In evaluating such expressions numerically, significant figures may be lost if the arguments of the two error functions differ only slightly, and if the arguments are both large and different. The latter case is particularly troublesome when the exponential factor multiplying the difference is large. To cope with these problems, program DELERF has been written for use on the Univac 1107. The program determines whether significant figures would be lost in a direct evaluation, and, if necessary, evaluates the difference by using a power series or continued fraction expansion.

RBU Evaluation. A correlation of experimental lattice parameters for a 19-rod cluster D₂O cell was made using the RBU Monte Carlo Code. A statistical analysis code was written and statistics for the calculated parameters determined. This completes the evaluation phase of the RBU development program. A document describing the results of the RBU test calculations is being written.

Cycle Analysis

Transient Behavior Studies for Fuel Survey

The rough draft of the paper "Dynamic Effects of Plutonium Fueling" is about 80% complete. The data appear good, and the observed effects are explainable in relatively simple terms. The principal conclusion will be that in many relevant situations the effective delayed neutron fraction will be sufficiently close to that for uranium fueling that concern over this effect from a fuel cycle analysis cost point of view is not greatly justified.

This seems to be true, for example, for the case of recycle level plutonium in enriched uranium for the case of graded fuel operation. This, however, assumes that heterogeneous effects in both the space and time variables are not too important, but current thoughts are that this is the case.

Thus, the computer package developed for this study will now be used to "check out" the reactor operating modes identified by the fuel survey code, ALTHAEA. To date, ALTHAEA runs are not investigated from this view point.

Reactor Engineering Development

Thermal Hydraulic Consequences of Primary System Debris

The possibilities of fuel overheating resulting from pieces of a rupture disc lodged in PRTR fuel elements or inlet piping components were considered. A recent examination showed that during the current shutdown period a piece had broken out of the rupture disc in the primary system block valve. Particles

of this disc had been carried throughout the primary system. Although efforts are being made to recover the missing pieces, by recirculating the coolant through screens, it is improbable that these efforts will be completely successful. Seven flushes have resulted in the collection of about 50% of the 2-1/2-inch diameter piece broken from the rupture disc; however, the amount collected per flush is decreasing, the seventh flush produced only about 1/2 of 1%. There is some possibility that pieces will become lodged in fuel elements or process tube inlet piping components during future reactor operation. Resulting flow disturbances could conceivably produce abnormally high fuel element temperatures.

The present situation has similarities to the event occurring several years ago in which pieces of a coffee can were spread through the primary system. Therefore, the earlier conclusions were reviewed for applicability to the present situation. There are two primary areas of concern:

- particles lodging in inlet piping restricting flow to a fuel element, and
- particles partially plugging flow paths through a fuel bundle, with local overheating.

In the present situation, no serious consequences should result from a partial restriction of inlet piping components. Pieces of the rupture disc must pass through the steam generator tubes to travel from the block valve to the reactor. The steam generator tubes have smaller diameters than the flow venturis, the smallest diameter components in the reactor inlet piping. A particle which could pass through a steam generator tube would not plug inlet piping components completely. Partial plugging could occur if an irregularly shaped particle lodged crosswise in a venturi or orifice. However, if this resulted in a substantial flow reduction, the reactor would be shut down by automatic safety circuit trips, and no fuel damage would occur.

It is possible for particles small enough to pass through the steam generator tubes to lodge between rods in a fuel bundle. Complete flow blockage would not result, since water could flow into the plugged channel downstream of the point of the blockage through spaces between the fuel rods. However, a relatively stagnant region could be produced immediately downstream of the particle. This could result in formation of a steam pocket and local high fuel surface temperatures. Although maximum specific fuel powers with the batch core will be about 80% higher than with the full length core being irradiated previously, the conclusions drawn for the earlier, "coffee-can" incident are valid for the present case. The blockage would not remove coolant flow over the entire circumference of a fuel rod and heat would be conducted away from the steam blanketed area, through the rod, to cooled surfaces. Temperatures would not reach the melting point of the Zircaloy cladding but could be high enough to result in a cladding rupture.

It should be noted that particles of foreign materials could be in the primary coolant system at any time. Damage to sealing gaskets, fuel bundle binding bands, and other components could produce such particles. Thus, the

present situation is not unique. The difference is that some amount of material is known to be loose in the system. Therefore, the probability of problems associated with such material is higher than usual.

Boiling Burnout Limits for FERTF Fuel Elements

Studies are being conducted to determine operating limits which will prevent boiling burnout in fuel elements to be irradiated in the PRTR Fuel Element Rupture Test Facility. The proposed fuel assembly will consist of eight 0.565-inch diameter fuel rods spaced uniformly in the annulus between a central, unheated, 0.983-inch diameter cylinder and an outer 2.503-inch ID basket tube. The assembly will be placed inside the FERTF pressure tube in the reactor.

No experimental burnout data exist for fuel assemblies of this configuration, a ring of fuel rods in an annulus. However, large numbers of experiments have been conducted for bundles of fuel rods inside a tube and for annuli with one wall heated and one wall unheated. Such data, although not specifically applicable, should provide guidance in setting adequate operating limits for the FERTF elements. Documents containing burnout data for rod bundles and annuli are being searched to obtain information for the pressure, heat fluxes, flow rates, and dimensions of interest for the FERTF fuel element.

Rupture Loop Particle Separator

Flow tests of the flow separator outlet nozzle at 314 Building indicate that the jet issuing from the nozzle may cause damage to the downstream elbow. Design of loop inlet and outlet piping, connecting to the separator, is being modified to provide a minimum of 40" of straight pipe run downstream of the nozzle (requirement based on 314 Building tests). Installation of the separator is planned for February, pending satisfactory completion of tests by Equipment Development.

Fabrication of the test loop by Equipment Development was completed, and preliminary calibration runs on the loop have been completed using a dummy test section. Fabrication of the separator is estimated 98% complete (delivery is scheduled for January 30). Testing will start as soon as the separator is received and installed.

Fuel Element Testing

Fretting studies of the wire-bound fuel element were resumed. To date, the element has undergone 535 hours of testing at 155 gpm, 1075 psig, and 525 F. During this period it has been removed three times for inspection by representatives of Fuels Development.

Materials Development

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 involves the determination of a hydrogen limit on unirradiated tube specimens containing flaws of various lengths and crack propagation tests at various temperatures. The respective objectives are to determine hydrogen and crack length limits for safe operation.

The 300 C burst stress of a specimen from discharged PRTR pressure tube 5696 was 81,000 psi at a calculated fluence of 2×10^{21} , $E > 1$ MeV. This stress represents a continuing and expected increase in strength with exposure.

Another experimental point obtained on a plot of failure strength of a pressure tube section vs test temperature reveals a -104 psi/ $^{\circ}\text{C}$ linear decrease in strength with temperature up to at least 300 C. Previous data up to 150 C was extrapolated to give a 300 C burst stress of 37,000 psi compared to the measured 300 C burst stress of 38,600 psi.

Ceramic Fuel Dissolution Studies

Ceramic fuel dissolution work during the past month has been centered on determining the influence of fluid flow on the dissolution rates of sintered UO_2 in OPG (oxalic-peroxide-fluconic) decontamination solution and HNO_3 .

Dissolution measurements on sintered UO_2 OPG solutions under quiescent conditions provided data for an Arrhenius plot. The next phase of defining the behavior of the OPG process was to determine the effects of flow on dissolution. A flow of 3 ft/sec at 50 C across the pellets increased dissolution by a factor of 2.4 over that found under static conditions. Measurements at other flow rates also produced data showing increased dissolution. Taylor and his co-workers in "Dissolution Rates of Uranium Dioxide Sintered Pellets in Nitric Acid Systems," Journal of Applied Chemistry, Vol. 13, January 1963, reported that flow decreased the dissolution rates on UO_2 . Experiments in our laboratory show that UO_2 dissolution in 4N HNO_3 is decreased by a factor of 10 by increasing the flow past the pellet from 1 ft/sec to 3 ft/sec. Reaction products apparently catalyze HNO_3 dissolution of UO_2 . Increases in flow result in lower reaction product concentrations at the surface which lower the reaction rate. The OPG reaction mechanism is the simple dissolution of UO_2 where reducing the reaction product concentration or alternatively transporting reactants to the surface accelerates the reaction.

P-4 Valve Rupture Disc

Examination of the P-4 valve rupture disc revealed the presence of well oxidized, and also fresh fracture surfaces, indicating failure prior to the previous system filming of PRTR. Cracking was associated only with the rupture

area, indicating a burst type failure followed by a fatigue breakup of the remaining exposed pieces of the rupture disc. Alpha activity contamination of the crevice area (associated with the assembled valve and rupture disc) was $\sim 100,000$ d/m.

Primary System Filtering

The primary system has been flushed through fine screens before startup of the reactor to remove any debris which was inadvertently introduced into the system during routine maintenance operations. The major portion of this debris was ion exchange resin and flakes of metal and corrosion products. Several other unidentifiable materials have been submitted for emission spectrographic analysis.

Postirradiation Examination of PRTR Zircaloy Corrosion Specimens

Sixty Zircaloy-2 and Zircaloy-4 tubular corrosion specimens were arranged on a rod and exposed in the PRTR at a position normally occupied by a fuel rod. The specimens were exposed in pH-10 lithium hydroxide, <0.14 ppm O_2 , in $>96\%$ D_2O at 246 to 279 C (475 to 532 F) for 209 days. The exposure was 231.6 MWd; the estimated maximum fluence was 5.7×10^{20} n/cm²sec, >12 keV. The tubular Zircaloy-2 and Zircaloy-4 specimens were 0.505-0.510 inch ID, 0.559-0.576 inch OD, and 1.000 ± 0.005 inch long. The specimens were exposed in three surface conditions: as-etched, autoclaved to prebreakaway weight gains*, and to post-breakaway weight gains.** Each type of sample appeared on the rod at ten positions, providing comparisons of oxidation as a function of flux level and temperature. Identical tubular specimens were exposed out-of-reactor in an autoclave in pH-10 LiOH in deoxygenated H_2O at 275 C. The specimens were decontaminated in an oxalic acid ammonium citrate decontaminant. Summarizing the results of the test:

- For each alloy and surface treatment, the in-flux weight gains follow the flux profile, indicating that flux accelerated the oxidation.
- On etched specimens, the effect of flux was slight, only factors of 1.2 greater than out-of-reactor weight gains at the highest flux. At the low temperature, low flux positions, weight gains were consistently less than the corresponding out-of-reactor weight gains.
- On specimens with prebreakaway autoclave films, weight gains were markedly affected by flux. Weight gain ratios (in-flux/out-of-flux) were 4.0 to 6.2 for Zircaloy-2 and 2.0 to 4.8 for Zircaloy-4.

* 20 hrs in steam at 400 C (750 F) and 1500 psi (17.4-19.6 mg/dm²).

**644 hrs in steam at 425 C (797 F) and 1500 psi (57.0-72.0 mg/dm²).

- In-flux weight gains on specimens with postbreakaway films were slightly lower than weight gains on specimens with prebreakaway films. However, in-flux/out-of-flux ratios were much smaller (up to 1.5 for Zircaloy-2 and Zircaloy-4) due to relatively large weight gains on out-of-flux specimens. The large out-of-flux weight gains are likely the result of a "memory" phenomenon from the 425 C postbreakaway autoclave treatment.

The in-reactor weight gains are in general agreement with results on six zirconium alloys exposed in the Engineering Test Reactor in pH-10 NH_4OH , <0.05 ppm oxygen in etched and prefilmmed (400 C [750 F], 1500 psi steam) conditions (A. B. Johnson, Jr., BNWL-CC-761, and BNWL-SA-822).

Hydrogen analyses will be performed on selected in-reactor and out-of-reactor specimens from the PRTR test.

Test Reactor Operation

Operating Experience

Approval to operate the PRTR was received on January 11, 1967.

On January 3, an inspection of the fuel elements and the rupture disc on the 14" motor-operated gate valve (P-4) indicated the need to flush the primary system and recover parts of the mechanically defected rupture disc and other maintenance debris. Cone screens, installed in all 84 process tube nozzle outlet openings, were used, and 14 separate flushes were instituted. The cleanup of the primary coolant system and the routine and special pre-operational and startup checks were completed and criticality achieved at 0200 on January 25, 1967.

All fuel elements which had been in the PRTR were returned to the 308 Building for inspection, and 55 fuel elements were recharged into the PRTR.

Batch Core power tests are in progress at month end.

Process Technology

Critical tests (14-2 and 14-3) were completed with the six-rod FERTF fuel element in rings 4 and 5 and with a mockup of the eight-rod FERTR fuel element in ring 0 (process channel 1550).

Batch core flux peaking measurements continued. A radial peaking factor of 1.16 was observed for an HPDC fuel element which was D_2O cooled in process channel 1550.

Complete pre-startup radiation measurements were obtained from pre-selected spots on the primary coolant system and the moderator and dry gas systems. Reactor operation, to 2 MW, did not reveal any unexpected increase in the radiation readings.

The solid material collected in the 5-micron crud filters during and after the primary system prefilming was analyzed as mostly gamma activity. An alpha to gamma ratio was about 1/100 d/m.

The filters have been removed from the primary system because of insufficient flow through the filters.

Three new corrosion measuring probes were installed to determine the corrosion rate in the biological and thermal shield coolant systems. Initial corrosion rates indicated 0.1 to 0.2 mil per month as expected. Corrosion probes in the moderator, reflector, and top and bottom shield cooling systems continue to indicate very low corrosion rates of less than 0.05 mil per year.

Analysis of samples taken during the decontamination of the FERTF showed equal quantities of plutonium in filtered and unfiltered samples and nearly equal ratios of plutonium to uranium. These results substantiate earlier indications that the decontaminating solutions effectively removed plutonium as well as uranium residues.

Improvement Work Status

Work Completed:

DC-226, Rev. 1. An arc suppressing capacitor and low burden (high resistance) coils have been installed on certain safety circuit relays to reduce the amount of current to be interrupted by relay contacts. This will reduce arc damage to the contacts.

DC-279, Rev. 1. The piping of the deaerator and flash tank overflows have been modified to provide for better control of effluent streams. This was a part of project BCP-007.

DC-283. The readout instrument for the containment vessel air monitor has been moved to the control room to permit range changes even though vessel entry is impossible. This was a part of project BCP-007.

DC-293. A diaphragm operated valve, operated from the control room, has replaced the original manual valve on the IX-1 vent system. This will permit venting even though vessel entry is impossible and will help re-establish primary flow should it be lost. This was a part of project BCP-007.

NPC-417. Bellows were found broken on several piping penetrations in the LAS thermal barrier. Repairs were made using foaming silicone rubber in the bellows cavity.

NPC-418. The seat and plug of valve H-48 has been modified to include a nonmetallic sealing surface in an effort to reduce chronic leakage. Initial operation has been quite successful.

The backup emergency power line from 384 Building was found with low resistance to ground in a preventive maintenance check in mid-summer. The underground line has now been replaced by a new line and is fully operable. Part of the new line is above ground.

Design Work Completed:

DC-280. Installation design has been completed for new PRTR exhaust air monitors which are duplicates of the present effluent monitors.

NPC-415. An expanded range will be provided for the conductivity readout instrument in the range where the primary system will be operating at neutral pH.

NPC-416. A deep immersion pH cell will be installed in the primary system to correct part of the deficiencies of the present system.

NPC-420. Test drains will be installed downstream of certain containment valves where it is not currently possible to detect leakage during leak testing.

Work Partially Completed:

New Instrument Power Supply
FERTF Delta P Tap Relocation
RL Annex Ventilation Enclosure
Remote D₂O Addition Station
RL Bypass
RL Particle Removal Equipment
Remote Readout for Primary System
TW Tank Bypass
Battery Room Air Exhaust
Drip Cover for MCC-4
TW Tank Discharge Line Heater Wiring
New A- and B-Cell Sump Pumps
Remote Venting of Primary Pumps
Pressurizer Level Control

Design Work Partially Completed:

Project BCP-013 - PRTR Decontamination and Waste Control
Analysis of HX-1 and Primary Pump Supports
New Storage Basin Tray
Fire Protection Improvements
Dual Safety Circuit
Dual Containment Valves
Dual Fog Spray Control Valve.

Projects - Technical Guidance

CBB-001 - Increased Power Level. Deferred phase I items (moderator heat exchanger, boiler feed pump, and HX-1 relief) are still under review. The deaerator modifications await third party code review of the methods to be used in the cutting and rewelding of this coded pressure vessel (ASME Unfired Pressure Vessel Code - Section VIII).

BCP-007 - PRTR Waste Handling. This project is to provide improved waste handling capability and control of reactor systems during accident conditions. Design is complete, and over-all construction is estimated to be 60% complete.

BCP-013 - PRTR Contamination and Waste Control. This \$65,000 project is to provide improved contamination and waste control during accident situations at PRTR. Over-all design is estimated to be 50% complete.

There have been no further developments concerning our request to the AEC for an additional \$5,000 to initiate procurement.

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 AEC is preparing a bid package which includes 326 and 327 Building only. The 309 Building is not included because of limited funds.

EBWR Demonstration Program

Irradiation Testing of EBWR Prototype Fuel Rods

The latest burnup estimates for the two remaining capsules in the MTR are: Capsule GEH-14-523; 26,300 MWd/ton of fuel (6.5×10^{20} fissions/cm³) and Capsule GEH-14-524; 24,900 MWd/ton of fuel (6.2×10^{20} fissions/cm³). These capsules (the last of a group of 32) were fabricated with short pieces of EBWR, Zircaloy-clad, vibrationally compacted UO₂ - 1.5 wt% PuO₂ fuel rods (1.07 cm OD).

Examination of another capsule (GEH-14-529) is continuing. The burnup analysis indicates that the burnup as determined by radiochemical analysis is 18,200 MWd/ton of fuel (4.52×10^{20} fissions/cm³) as compared to a previously reported burnup of 22,100 MWd/ton. From flux monitor wires, it appears that the capsule operated at linear powers up to 17.4 kW/ft (571 W/cm) during the first 35.1 days. The capsule was shifted to a lower flux position for the remainder of the irradiation, 703.3 days, and operated at a maximum linear power of 12.1 kW/ft (397 W/cm). The capsule fuel released 14% of the fission gas.

Examination of the two production-run EBWR fuel rods is continuing. Burnup analyses show that the previous estimated values were in error. The analytical data indicate burnups of 5760 MWd/ton of fuel (1.5×10^{20} fissions/cm³) and 9220 MWd/ton of fuel (2.4×10^{20} fissions/cm³). No neutron radiography work was done.

The 21-rod bundle (Fuel Element No. 6501) with production-run rods is scheduled for recharging (after power tests) into the PRTR core periphery. The PRTR is presently undergoing power tests, prior to resuming normal operation.

A summary of the above capsule and rod tests is being prepared for submission to the American Nuclear Society for possible presentation at the meeting in San Diego next June.

EBWR Experiment

A trip was made to Argonne National Laboratory to discuss details of the EBWR experiment program to be conducted during the next six months. A report is being written concerning these details.

Irradiated Fuel Shipment

A study was made by the Nuclear Safety Specialist to determine if the PRTR cask could be licensed for inter-state shipments of irradiated fuel. It was concluded from this study that the cask could not be approved for this purpose and used for EBWR fuel shipments. For this reason arrangements have been made to use the Yankee single element cask for these shipments. The necessary arrangements for shipping are being made, and the first shipment of rods from ANL to PNL should be made this spring.

NUCLEAR SAFETY

Containment Systems Experiment (J. M. Batch)

Experimental Facilities

The reactor simulator vessel was placed in position in the 277-T Building test stand and installation of braces and pipe jumpers started. Conduit between the blowdown facilities and the control room in 221-T was installed in preparation for pulling the wires for process control and experimental sensors transmission systems.

Design is proceeding on a modified version of the Phase I dummy core for the reactor simulator vessel. It is being designed with arrangement flexibility so that it can be used initially as a simple resistance to flow, but later it can have additional parts added in the field to allow simulation of typical coolant flow configurations of pressurized or boiling water reactors. An analysis is being made of the loadings that may be expected on this dummy core during the subcooled depressurization portion of the blow-down tests.

Aerosol Studies

Run 78 was made in the Aerosol Development Facility (ADF) stainless steel containment vessel using a three-component aerosol mixture of iodine, cesium, and UO₂-Zircaloy fume. The containment atmosphere was steam-air, 80 C, 0 psig, isothermal. The gas phase concentration half lives were 10 ± 2 min for iodine, 10 ± 1 min for cesium, and 15 min for uranium. These values correspond to gas phase mass transfer coefficients of 0.020, 0.020, and 0.0133 cm/sec, respectively; values consistent with previous tests using one or two components. Ratios of cesium to iodine in various gas and liquid samples varied with sample type and with time, indicating that they had not interacted significantly. The diffusio-phoresis contribution of condensing steam was 0.0070 cm/sec, showing this to be an important but not dominating mechanism for mass transfer in a condensing steam environment.

A Maypack cluster assembly with Maypacks which will be used in the forthcoming large-scale CSE aerosol transport tests was checked out during this run. The cluster functioned properly, but two defects were found in the Maypacks. Some bypassing of the filters occurred because of insufficient gasket sealing pressure and the teflon balls in the inlet check valves disturbed flow patterns enough to cause excessive deposition on the interior of the nose cones. The first defect has been corrected; the latter will be difficult to solve completely.

Retention of elemental iodine on Maypack silver surfaces was studied. Trapping efficiency in room temperature air was shown to be poorer than in warm air or in steam-air. Three silver membranes with 5-micron pores were superior to six silver-plated copper screens of 100 mesh size. The decision was made to use six screens followed by one silver membrane in the CSE Maypacks. Efficiency for retaining elemental iodine will be >99% for any CSE sampling condition envisioned.

Decontamination efforts on coupons exposed during ADV Run 78 showed that cesium was easily decontaminated from both SS-304 and carbon steel painted with Phenoline 302,¹ but as in past efforts, iodine was not removed to any great extent from the paint by any reagent tried. The best decontamination was given by 1.0M NaOH at 95 C with cesium DF of ~500 for both materials, and iodine DF of 1000 for SS-304, but only 2.8 for Phenoline 302.

¹Phenoline 302 is a modified phenolic coating manufactured by the Carboline Company, St. Louis, Missouri.

A mathematical analysis of the behavior of fission products in containment systems was made by means of over-all material balances for the gas and liquid phases and on solid surfaces. Mathematical expressions were derived for time-dependent concentrations in the gas phase, in the liquid film, and in the condensate pool, and for the amount reacted with the vessel interior surfaces. A computer program was written for the PDP-7 computer to assist in correlating experimental data with theory. Input information required includes vessel volume and surface area, liquid holdup volume, steam condensation rate, gas phase mass transport coefficient (K_G), liquid-solid reaction rate constant (K_L), Henry's law constant, and a proportionality factor for relating concentrations in the wall film and the condensate sump. Outputs are the roots of the equations and tabulated values of dimensionless concentration versus time. Input values of the unknown variables can be adjusted by trial and error until the output agrees with experimental data from the ADF. The model fits the ADF experiments with high iodine concentration quite well. At lower concentrations, the model fits the general shape of experimental data, but departs significantly at extended times. This may result from loss of experimental accuracy at very low concentrations or some of the variables may be concentration dependent.

Analytical Data Acquisition

Final analyses for Run 78 (a mixed cesium-iodine aerosol test in the ADF) were completed using the on-line computer gamma energy analytical system. One-hundred-seventy individual samples were counted using two of the four detector systems. The majority of run samples was processed in two shifts, and a fraction of this time was spent in assuring proper instrument performance for the first actual analysis of run samples. Successive counts on the two detector systems were within statistical limits and were within 2% agreement for those samples counted on both detector systems. Cesium recovery for the total number of samples taken was 95% while iodine recovery was 86% of input. Extrapolation of deposition coupon data to the release vessel internal area (painted Phenoline 302) can account for the observed low iodine recovery.

Leakage Rate Testing

Leakage testing continued. A 30-psig and two 10-psig tests were completed at a nominal 1%/day leakage rate condition. A consolidation of the test data appears below:

<u>Dates & Comments</u>	<u>Ref. Method %/day</u>	<u>Abs. Method %/day</u>	<u>Collected Leakage %/day</u>
30 psig, 1% 3-day average 12-10 to 12-13	0.63	0.66	0.55
10 psig, 1% 7-day average 12-13 to 12-20	0.28	0.38	0.28
10 psig, 1% repressurize & repeat 8-day avg. 12-21 to 12-29	0.31	0.38	0.28

The 1%/day nominal leakage rate condition means that the containment vessel leakage rate was established at about 1%/day at a pressure of about 60 psig; and leakage rate tests are then performed at a series of decreasing pressures without disturbing any of the leakage points.

A typewriter coupled digital voltmeter is being used with a programmed timer to gather temperature and pressure data over 24-hr periods.

Instrumentation Development

An ultrasonic anemometer scheme has been demonstrated in room temperature air. Continuous wave ultrasonic beams driven in opposite directions are superimposed one on the other electronically, and the phase shift caused by medium velocity (air) is converted to voltage to drive a recorder. The configuration is sensitive to room air currents. The absolute sensitivity will be determined in calibration tests. The instrument will be mounted on an ADF tank for a test in a steam and air containment atmosphere.

Preparations are being made to experimentally demonstrate the performance of the neutron densitometer using certain CSE blow-down test components. The pipe to be used in the experiments, a spool section of schedule 160 steel pipe, is available. The water fraction calibration standard, now partially fabricated, will be completed as soon as possible. This standard has a honeycomb configuration of 80 small diameter aluminum tubes that will be filled to yield varied water fractions and geometry conditions.

Pressure Vessel Crack Monitoring (J. C. Spanner)

Detection of Metal Overstress by Acoustic Emission

During the past month the primary emphasis in the program to apply acoustic emission in metals as the basis for a crack monitor system for surveillance of reactor pressure piping was directed toward compiling the second quarter FY-67 progress report. The report was completed and is ready for inclusion into the PNL Nuclear Safety quarterly report.

Specimens are being prepared for a preliminary study of acoustic emission generated by flaw growth in internally pressurized vessels. The specimens are being fabricated from three-foot long sections of three-inch diameter, 0.250-inch wall A-106 carbon steel pipe. These specimens will be caused to fail by static internal pressurization with cold water. This series of tests is the planned predecessor of a test to demonstrate detection of acoustic emission under dynamic conditions in a simulated field application.

Arrangements have been made with Mechanical Metallurgy Unit to participate in the fracture testing of heavy plate specimens (1/2" thick) to investigate and analyze the accompanying acoustic emission. This provides an opportunity for

initial comparison of acoustic emission in thick vs thin specimens and to correlate acoustic emission signals with the metallurgical sequence of metal fracture.

Two new charge amplifiers have been constructed employing a low noise field effect transistor (FET) as the first stage of amplification. The noise level of these units is about one-tenth that of the commercial bipolar charge amplifiers used in previous acoustic emission work. A prototype of these FET amplifiers has been used successfully with accelerometers on double cantilever beam specimens. The inherent low noise of these amplifiers allows better definition of the continuous frequency phenomena which is an important part of the emission signals.

The response characteristics of the following four charge amplifiers were evaluated for applicability to acoustic emission work: (1) a commercial FET amplifier, (2) two types of commercial bipolar transistor amplifiers, and (3) the locally fabricated FET amplifier. Test results indicated the superiority in noise level characteristics of the FET amplifiers, but the locally constructed FET model lacked the bandwidth of the commercial bipolar transistor amplifiers. The commercial FET amplifier exceeded both the low noise and bandwidth capabilities of the bipolar units. All of the amplifiers tested, however, were capable of passing frequencies to 25 kHz. At approximately this frequency, accelerometer resonance becomes important, and the rolloff of the amplifier is compensated for by an increase in transducer sensitivity. Since accelerometers are the principal transducer used in present testing, the commercial FET charge amplifier would offer no advantages over the locally fabricated ones. However, when the electrostatic transducer is available for acoustic emission tests, the 150 kHz bandwidth of the commercial FET amplifier will be desirable.

An electrostatic transducer concept is being developed for use both as a wideband nonresonant detection for acoustic emission and, also, as a point source shock pulser (i.e., broadband pulse) for simulating crack propagation signals. Preliminary results indicate the transducer operates as anticipated, and no resonances were detected from 1 Hz through 1 MHz although signal amplitude decreases with increasing frequency above 10 kHz.

Reactor Safety Analysis and Evaluation (R. G. Wheeler)

One portion of the program is concerned with evaluating the failure behavior of irradiated low alloy steel pressure vessels from tests on subsize vessel specimens. The results of the subsize vessel evaluation work are currently being prepared for presentation in a formal report.

A second phase of the program is the testing of irradiated and unirradiated larger models and large fracture mechanics specimens. One of the initial problems of this work has been finding suitable reactor facilities for irradiating large specimens. A potential irradiation facility is the Sandia Experimental Reactor Facility (SERF) in Albuquerque, New Mexico. Although the neutron

environment of SERF is relatively well established, the amount of gamma heating to be expected with large section specimens is not adequately known. During this period an experiment was prepared and shipped to Sandia for irradiation in the SERF. This experiment will yield the information required for predicting the gamma heating of the large section specimens irradiated in the SERF.

Waste Solidification Condensate Treatment (B. W. Mercer)

Laboratory experiments were conducted to evaluate the effectiveness of waste incorporation in asphalt as a method for treating glass melter condensate. Asphalt products containing 60 wt% waste solids were prepared from neutralized, simulated, glass melter condensate by the method developed at ORNL. Neutralized condensate was introduced into emulsified asphalt (Type RS-2: a rapid-setting, high viscosity, asphalt containing 63 wt% asphalt, 35 wt% water, and 2 wt% emulsifying agent) at 65 C, and the water was evaporated by heating to 160 C. The mixture was stirred continuously at 150 RPM by a four-bladed paddle that scraped the walls of an electrically heated stainless steel vessel. Free flowing asphalt products were prepared from both neutralized condensate and from condensate that had been previously scavenged with phosphotungstic acid and MnO_2 (pH 12) to reduce fission product activity. The latter contained sufficient Cs-137 for static leach tests which are conducted by contacting the asphalt product with distilled water at 25 C. The water is sampled and renewed each week.

The cesium-137 leach rate was $7 \times 10^{-5} \text{ g cm}^{-2} \text{ d}^{-1}$ after four weeks. This leach rate is higher than that determined after one week; however, it is believed that the first two samples were low due to inadequate mixing before sampling.

Fission Product Aerosol Containment

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

Additional data were obtained on drop size and spray patterns for nozzles in the 4 m³ chamber to be used in the hydrazine study. Drop size and spray patterns are of importance in the interpretation of the spray chamber results based on the kinetic data obtained and mass transport considerations. All nozzles studied yielded larger droplet diameters than anticipated, and a very significant fraction of the total flow impinged on the walls of the four-foot diameter chamber. Although mass transport of methyl iodide to droplets of about 80 μ diameter was indicated not to be the rate limiting process in the removal of sprays, the simplifying assumptions of a smaller, uniform drop size, and no wall effects limit the validity of the conclusion, and a more realistic estimate will be required. For example, a preliminary calculation suggests that the wetted surfaces may be more significant than the droplets themselves in removal. Provisions are made in the design to determine the significance of the wetted walls.

The spray nozzle experiments were followed with heating performance tests of the spray chamber. Various details were completed, the iodine generator calibrated, and the plastic enclosure installed.

$$\left(\frac{\text{activity leached/cm}^2 \text{ d}}{\text{activity/g of product}} \right)$$

Physical Chemistry of Hydrazine-Methyl Iodide Reaction (L. L. Burger)

The results of studies on the heterogeneous reaction between hydrazine and methyl iodide in a static system described earlier indicate that the rate of change of methyl iodide is proportional to the interfacial area, mole fraction of $\text{N}_2\text{H}_5\text{OH}$ in the liquid, CH_3I concentration in the gas phase, and the reciprocal of the volume of the gas phase.

To test the validity of these conclusions on the rate of change of methyl iodide, a gas washing system was designed which would increase the ratio of interfacial area and gas volume. In the experiments the bubble size and contact time were estimated through visual observation while the experiment was in progress; hence, only approximate results were achieved. Small bubbles of about 1 mm diameter were obtained by passing the gas through a glass frit into the aqueous alkaline hydrazine solution. The height of the solution was 3 cm, giving a contact time of about 1 second.

Iodine-131 labeled methyl iodide was prepared by the iodide-methyl iodide exchange reaction. The exchange solution was diluted by a factor of 20, and the methyl iodide was then transported into the scrubbing units by a flow of air through the system. A series of three scrubbers followed the methyl iodide generating vessel.

A flow of carrier gas of either 10 ml/min or 30 ml/min was continued for 100 minutes after which the various solutions were sampled. The total activity of I-131 in the solutions was determined.

The total methyl iodide entering the first scrubber was assumed to be the total found in the three scrubbers in series. The concentration of hydrazine was 33.7 wt%. In 100 minutes of sparging, 96.5% of methyl iodide was retained in the first scrubber when the flow was about 10 ml/min, and 89% was retained in the first scrubber when the flow was 30 ml/min. About 6% was trapped when water was used.

Over-all reaction velocity constants were calculated. Static experiments had shown that the rate of removal was proportional to exposed surface area and to the reciprocal of volume. Comparing static to scrubbing tests, one should find

$$\frac{k_2 V_2}{A_2} = \frac{k_1 V_1}{A_1} ,$$

where the subscripts 1 and 2 refer to static and scrubber tests, respectively, A is the area of the interface and V is the gas volume contacting the interface.

The scrubber reaction rate constant should then be given by

$$k_2 = \frac{k_1 V_1 A_2}{A_1 V_2}$$

The observed k_2 was about four to six times as great as that predicted from the static tests. With the uncertainties in estimating the parameters, this agreement is considered adequate to help support observations on the static tests.

The rate at which methyl iodide exchanges with iodide ion was determined to be about 0.03 per hour, equivalent to a bimolecular rate constant of $6.1 \times 10^{-5} \text{ mole}^{-1} \text{ sec}^{-1}$, a value much lower than that reported in AERE-R-4823.

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

The single well solution for injecting gas into homogeneous soils provides an economical method for determining the flow rates and minimum times of travel back to ground surface for a wide variety of geometry factors, injection pressure, and temperature. Graphs have been prepared for dimensionless flow rates as functions of dimensionless geometry factors. From these graphs, which are independent of pressure and temperature, the flow rate in standard CFM is easily obtained. Similar graphs for minimum travel times are now being prepared. Results can be used for determining the optimum placement of the injection structure at a given site or for assessing the concept of disposal for a wide range of possible field sites. These results will be used to help design site evaluation tests at the NRTS and to choose permeability values for sediments overlying the basalt.

Geophysical Exploration of Rattlesnake #1 Well (W. A. Haney)

The Rattlesnake Hills Test Well Evaluation Project writeup was completed by BNW Facilities and Engineering personnel, and bid packages are being assembled by BNW Legal personnel. Entry and work permission was given by the landowner. Proposed work was discussed with the Division Geologist, Western Operations, Standard Oil Company of California; the State Geology Dept.; and the Tacoma office of the U.S.G.S. Standard Oil Company opened their file on the well for our examination and will supply copies of logs and other information that were not released to the public. They are also sending drilling samples for our examination. A portion of the vertical core taken in the well at 8100 ft was received from the State Geology supervisor, and thin sections are being prepared from this sample. The core was described as "basalt" in the Standard Oil Company logs, but it appears to be a consolidated tuff from cursory examination. X-ray diffraction studies of a white mineral in the core showed it to be quartz, and diffraction studies of the ground mass of the core showed only trace quantities of two zeolite minerals. No other crystalline minerals were evident.

Columbia River Sedimentation Studies (D. R. Kalkwarf)

Analysis of data for a special report to the AEC on the effects of the reactor shutdown last summer was completed. The transport and deposition of five radionuclides, Mn-54, Co-60, Zn-65, Sc-46, and Cr-51, in the Columbia River were followed extensively, with the conclusion that substantial quantities of the first four nuclides were redissolved from bottom sediment in the river during the shutdown period. Two types of evidence led to this conclusion. Immediately after shutdown, the amounts of these nuclides injected into the river as reactor effluent dropped to between 1% and 10% of their previous values. Transport rates, measuring the movement of radioactive solutes and suspended sediment in the river, did not reflect this drop as sharply downstream and showed that more radioactive material passed the Pasco, Umatilla, and Vancouver stations than would be expected from the low reactor output. Measurements were also made on cores of bottom sediment which showed that the decrease in radionuclide content of the top inch of sediment exceeded that attributable to radioactive decay.

Radioactive Waste Solidification (A. M. Platt)Process Assistance

A sample of 324 Building demineralized water was examined after the B-Cell spray calciner filters plugged during pre-run testing in which the spray calciner was being operated on "demineralized" water only. The water was almost 0.3M in phosphoric acid. Filter plugging was probably due to high boiling phosphoric acids and was similar to plugging that has been observed previously when high phosphate feed solutions were processed.

In WSEP Runs 3 and 4, PW-2 will be processed by the Pot-Cal procedure. Purex 1WW waste procured for these runs is high in aluminum ion; consequently, WSEP feed prepared from this 1WW is more concentrated in aluminum than PW-2 solution. Candidate WSEP feed solutions for WSEP Runs 3 and 4 in which the iron concentration was below that in PW-2 to compensate for the excess aluminum were prepared and tested in the laboratory for foaming, sulfate retention, and for Mode C operation. Both feeds appeared satisfactory for processing by the Pot-Cal procedure. A series of laboratory tests was made to study Pot-Cal processing of PW-1 waste containing excess sodium and aluminum. Foaming is a problem with these solutions. The tendency to foam could be reduced significantly by addition of sulfate or lowering the acidity from seven to about four molar.

A Ni-Cr-W alloy developed by Owens-Corning Fiberglas Corporation (US 3,170,787) for fabrication of perforated plates used to throw out glass fibers at about 1100 C was evaluated in a PW-2 type melt (SCM 132, M⁺/P = 2.0) at 1200 C. The specimen was dissolved during a 24-hr test. Several ceramic materials (silicon carbide, silicon nitride, zirconium diboride, titanium carbide, and molybdenum disilicide) and two cermets (Haynes LT-1 and Haynes LT-1B) were tested in a PW-2 melt (CSCM-9) at 1000 C. Attack by the melt

ranged from very slight on silicon carbide, silicon nitride, zirconium diboride, and Haynes LT-1B to moderate on molybdenum disilicide to severe on titanium carbide and Haynes LT-1.

Product Evaluation

Preparations to measure leach rates on radioactive samples from initial WSEP runs are in progress. The first of these measurements will be started in a 325-B Building hot cell in early February. Several leach tests on nonradioactive simulated products from the spray solidifier processing of PW-1 (PNL:PW-1) and PW-2 (PNL:PW-2) were completed. Purpose of these tests was to obtain base line data for evaluation of results with radioactive WSEP samples. Results of several duplicate leach tests indicated good precision can be obtained with the procedure. Other tests showed that variations in the thermal history of a sample can cause differences in leach rates which exceed the experimental error in the procedure; thermal conditioning of WSEP samples prior to leach testing appears desirable.

The transducer for ultrasonic testing of the radioactive pots in WSEP failed. It was successfully replaced remotely, and the GE-412 computer was used to generate a thickness map of the receiver pot to be used in Run PC-2.

Radioactive Residue Process Development (A. M. Platt,
G. J. Alkire)

Waste Solidification Engineering Prototypes (WSEP)

The first radioactive spray solidification run was partially successful in meeting objectives. A 12-inch diameter container was filled 1/3 full of ceramic solid. Some 24 hours of operations were completed at 16 liters/hour without incident, but the run was terminated after 37 hours of feeding rather than complete a planned 100 hours. Apparently, poor atomization initiated buildup within the calciner. Early termination was chosen to prevent more serious accumulation. Acid flushes have been used to clean the calciner interior. The cause of poor atomization is being investigated by a series of nozzle tests.

The "sulfate-free" wastes received from the Purex plant was high in Na and Al concentrations from cross contamination by coating solutions. The planned PW-1 flowsheet runs were postponed for pot calcination, and the sulfate flowsheets are being tested. A new "uncontaminated" feed batch will be sought to provide pot calcination tests on the sulfate-free flowsheet.

GE-412 Computer Studies

The spray calcination data logging program failed to achieve a fully operational status in time for the 412 computer to satisfactorily monitor Run SC-1. The difficulties were traced to several causes: (1) an inadequate

pulse length was generated by the calciner furnace on-off switches; (2) actual furnace switches had higher contact resistance than that of the simulator switch used in debugging operations, causing too low a relay coil current; (3) a "contact bounce" condition triggered the computer interrupt program twice for a single interrupt. These problems were corrected at month end, and the system is now operational.

Spray Calciner Development

The fourth spray calciner run in a series involving the PW-1 flowsheet was completed during the month. The last run was for a 10-hr period, used steam atomization, and had an average feed rate of 20.1 liters/hr. A new thermocouple had been located externally on the cone wall directly below the calciner barrel prior to the run. This thermocouple showed a continual decrease in temperature during the run and was the only indication that the run was not going smoothly. Inspection of the calciner after the run revealed 6.6 kg of caked calcine located in the cone directly below the calciner barrel. A summary is currently being prepared which will describe the results of the four-run series.

Batch Melting in Waste Container

Several Sauereisen¹ cement coatings are being evaluated as protective coatings for mild steel containers. All cements tested adhere well to the steel. The coated mild steel samples will be tested for attack by molten waste.

The engineering scale test run has been postponed to April as a result of the more urgent need for hot cell support work.

Alternative flux compositions were tested by Materials and Process Chemistry personnel with the following results:

Waste Type	Flux (fm/liter waste on 100 gal/ton U basis)	Melting Point °C	Melt Description
PW-1	Colemanite 410 gm/liter	925	Homogeneous viscous melt, forms brown crystalline solid.
PW-2	Colemanite 320 gm/liter	950	2 phases formed--heavy phase would not flow at 950 C. Solid consisted of 25% light phase.
PW-1	Na ₂ SO ₄ 479 gm/liter	-	Only partially sintered at 950 C.
PW-2	Na ₂ SO ₄ 370 gm/liter	875	2 phase; 20% light phase.

¹Sauereisen Cements Co.

Melting points above 900 C are probably too high for batch melting, and compositions having lower melting points are being sought.

Advanced Melter Concepts

Several materials have been screened as candidates for melter materials. Materials and Chemistry personnel have qualitatively assessed the degree of attack by PW-2 phosphate melts (run CSCM-9 composition) on various materials and determined whether the materials were permeable to the melts. Silicon nitride, sintered alumina, and cermets containing alumina, chromium and/or tungsten and molybdenum resist attack and are not permeable. The cermets appear especially promising because they are far less fragile than ceramic materials.

Solids Storage Environmental Testing

The initial package for component fabrication of controlled environment test pods has been completed by the CPFF contractor. Further component shop orders have been cancelled, and the remainder of the funds will be used for pod assembly and installation. Experience to date indicates that unit cost of the pods will exceed the original \$10,000 estimate by 20-30%, but the cost will be covered by the contingency allotment.

Design Verification Tests

The platinum melter was returned to the fabricator to rectify the nozzle distortion and to repair the cracked flange of the nozzle for the off-gas line. Attention is being directed to evaporator tests until return of the melter. The melter liquid level probe is being redesigned in an attempt to eliminate repetitive failures.

Waste Solidification Condensate Treatment

Laboratory experiments were conducted to evaluate the effectiveness of waste incorporation in asphalt as a method for treating glass melter condensate. Asphalt products containing 60 wt% waste solids were prepared from neutralized, simulated, glass melter condensate by the method developed at ORNL. Neutralized condensate was introduced into emulsified asphalt (Type RS-2: a rapid setting, high viscosity asphalt containing 63 wt% asphalt, 35% water, and 2 wt% emulsifying agent) at 65 C and the water evaporated by heating to 160 C. The mixture was stirred continuously at 150 rpm by a four-bladed paddle that scraped the walls of an electrically heated stainless steel vessel. Free flowing asphalt products were prepared from both neutralized condensate and from condensate that had been previously scavenged with phosphotungstic acid and MnO₂ (pH 12) to reduce fission product activity. The latter contained sufficient Cs-137

for static leach tests which are conducted by contacting the asphalt product with distilled water at 25 C. The water is sampled and renewed each week. The Cs-137 leach rate¹ was $7 \times 10^{-5} \text{ g cm}^{-2} \text{ d}^{-1}$ after four weeks. This leach rate is higher than that determined after one week; however, it is believed the first two samples were low due to inadequate mixing before sampling.

1 $\frac{\text{activity leached/cm}^2 \text{d}}{\text{activity/g of product}} = \text{g cm}^{-2} \text{ d}^{-1}$

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