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

**DIVISION
OF
REACTOR DEVELOPMENT AND TECHNOLOGY
PROGRAMS**

AP LARRICK 31484 326		JAN 31 1968
W.D. Greenhalgh 30912 325		FEB 6 1968

AUGUST, 1967

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RICHLAND, WASHINGTON

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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

FOR JULY 1967

AEC DIVISION OF

REACTOR DEVELOPMENT AND TECHNOLOGY PROGRAMS

By

The Staff of Battelle-Northwest

S. L. Fawcett, Director

August 1967

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

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SUMMARY

CIVILIAN POWER REACTORS

Nuclear Systems and Concepts Analysis

LP system changes as suggested by the Systems Analysis Task Force to correct treatment of ThO_2 discharges, narrowed convergence criterion and improved interface with the report writer have been made and debugged. Several other changes suggested by the committee are being debugged. BNWL-429 describing operation of the PACTOLUS code has been issued.

A comparison of uranium metal fuels to urania fuels has shown an advantage for the metal of nearly 0.03 mill/kWh if exposures of 10,000 MWD/tonne can be obtained. Optimizing the design might show a greater cost advantage.

The fuel cycle analysis chain of computer codes has been made available for offsite use by AECL and Purdue University.

Power Reactor Design Studies and Evaluations

Burnup calculations using FCC and ORNL rather than Russian cross sections showed essentially no difference in initial k_{eff} needed to obtain 50,000 MWD/tonne.

The pressure drop external to the core is now put into the design simulation as a constant rather than as a fraction of the in-core pressure drop.

The reason for discrepancies in the D-1 in-core pressure drop as calculated by the PNL design simulation and Karlsruhe have been identified and are being evaluated.

Gross heat capacity calculations have been completed for the major streams in the South Atlantic Slope Basins, James River to Savannah River and the Eastern Gulf of Mexico Basins, Ogeechee River to Pearl River.

USAEC-AECL Cooperative Program

Tensile test results for unirradiated control specimens and pre-crept in-reactor creep specimens show no strengthening with in-reactor exposures at 350 and 400 °C. The yield stress was reduced as a result of the in-reactor exposure under stress.

Corrosion data are reported for six zirconium alloys exposed 174 days in-flux and out-of-flux in the Engineering Test Reactor G-7 loop. The in-flux corrosion resistance varied widely as a function of alloy.

HTGR Graphite

The radiation-induced water vapor-graphite reaction has been studied by attempting to observe the products of the reaction H_2 , CO , and CO_2 . To date, no products have been observed by an analysis with a detection limit of 3, 2, and 4 vpm for H_2 , CO , and CO_2 , respectively. Greater sensitivity will be required to determine the rate of the radiolysis reaction.

Analyses of small quantities of water vapor have been attempted by converting the water to hydrogen by reaction with calcium hydride or to acetylene by reaction with calcium carbide. The gaseous products are analyzed by gas chromatography. The use of calcium hydride was unsatisfactory because of the high blanks when no water was added. However, the use of calcium carbide looks promising.

APPLIED AND REACTOR PHYSICS

Plutonium Criticality Studies

Testing and calibration has proceeded on small spherical (3.9 cm dia.) proton recoil counters for use in neutron spectrum measurements. Preliminary measurements were made of the fast neutron leakage spectrum from an undermoderated heterogeneous plutonium assembly comprised of layers of PuO_2 -polystyrene and Lucite.

As a guide for planning critical experiments with mixtures of plutonium containing Pu-238, a series of calculations were completed for estimating criticality of material, for use in experiments, that contains about 18 wt% Pu-238, 65 wt% Pu-239, 10.4 wt% Pu-240, 5.4 wt% Pu-241, and 0.8 wt% Pu-242.

The latest version of the British Monte Carlo Code, GEM-3, is now operable on the 7090 computer. Criticality for a reflected assembly of PuO_2 -polystyrene in nearly cubic geometry was computed with good results.

Containment tests, required for safety, were undertaken to determine the ability of the reactor room to meet original acceptance tests. The leakage rate was found to be above the acceptable design limit; steps are being taken to improve the containment.

Phoenix Fuel Reactor Program

New Al-Pu fuel plates and test-assembly hardware were completed and shipped to the MTR for the irradiation experiment.

Preliminary fuel element and shim control rod specifications have been prepared and reviewed by Idaho Nuclear Corporation and BNW personnel.

Six hundred uranium oxide fuels, 12 cadmium poison follower rods, and 16 follower fuel sections are to be modified for a PRCF fuel experiment.

A summary entitled "Critical Experiments with Phoenix Fuel in an MTR Mockup," has been submitted for presentation of a paper at the 1967 Winter Meeting of the American Nuclear Society.

Calibration of one interior and one peripheral shim in the PRCF-Phoenix core has been completed.

A PRCF loading with fully inserted control rods amenable to 2-D analysis has been suggested.

Better agreement between calculated and experimental power profiles may be obtained by using THERMOS/BNW than by using THERMOS/HAMMER.

A document describing the 3-D codes WHIRLPOWER and POWERWRITER is in the final publication process.

The effect of variation in pressure head and variation of inlet coolant temperature on operating power limits is being studied for a Phoenix core in the MTR. Feasibility, from a thermal and hydraulic viewpoint, of increasing the number of fueled plates in the shim rod follower from 12 to 13 is under investigation.

A solid fuel monitor wand, which will be easier to fabricate and less susceptible to damage during handling, is being considered for application in the MTR Phoenix fuel experiment.

A number of heat transfer limited reactor core designs for a range of fuel volume fractions and pin diameters have been calculated for possible maritime application of the Phoenix concept. Enrichment requirements and material balance corresponding to these cases are being found for fuel cycle cost analysis input.

A listing of typical HFIR calculations has been received from ORNL.

High Temperature Reactor Physics Studies

The GAS CYCLE-HEAT design tests on the High Temperature Lattice Test Reactor (HTLTR) were stopped at a reactor temperature of 675 °C because of a failure of the control circuits. The reactor was then cooled.

Inspection of the reactor showed essentially no corrosion, but some graphite bolts were broken in tension. These have been replaced, and many minor modifications have been made to the reactor and its systems.

The vertical safety rods were modified as proposed. One was repaired after it was dropped during modification work.

The horizontal control rods were installed while inverse multiplication measurements were taken.

Preparations for NUCLEAR-ROD design tests are in progress.

Negotiations have been completed to obtain pyrolytic carbon-coated U-233 O₂ microspheres. These will be used in reactor loadings which are close to currently proposed high temperature graphite gas-cooled reactors.

REACTOR FUELS AND MATERIALS

Fast Fuels Oxides and Nitrides

Kinetic measurements on nitride synthesis via the carbo-thermic reduction of the oxide were made for UO₂ and UO₂-20 wt% PuO₂.

Experimental work was begun during the month using the 8-inch spray calciner to produce UO₂ - carbon mixtures starting with UNH-sugar solutions.

Two experimental mixed nitride fuel capsules continue to operate satisfactorily in the MTR after accumulating exposures of approximately 21,000 Mwd/tonne. Two previously irradiated capsules were returned to Hanford for postirradiation examination.

Basic Swelling Studies

Capsule P-10 (450 °C, 5000 psi, 0.2-0.8 at.% BU) was discharged after successfully reaching its goal exposure. Two other capsules continue to operate as designed, and construction is complete on three more capsules.

Specimens of uranium irradiated at low temperatures show much less swelling after a 100-hour anneal at 15,000 psi and 900 °C than they did after a similar anneal at low pressures. The high pressure treatment also caused densification of samples which had previously swelled due to high temperature irradiation.

Nondestructive Testing

A study has been initiated to determine the feasibility of using an automated system to adjust the parameter separating circuits of the multiparameter tester. Test circuits for applying the multiparameter test techniques to a pulsed eddy current system are being assembled, and a detailed analysis is being made of the functioning and adjustment of the parameter separating circuits of the multichannel eddy current tubing tester. The effect of test coil replacement and instrument drift on the reproducibility of instrument output is receiving special emphasis.

Ultrasonic, mechanical, and electronic apparatus has been assembled and steel specimens are being irradiated for the purpose of developing a test method to detect radiation induced shift in the nil-ductility-transition temperature of structural steels. Several electronic units have been integrated into a single package, and an RF gate, peak amplitude pulse stretcher, and other circuits are being developed to provide an output signal of maximum utility.

Analysis of ultrasonic wave propagation in an attenuation media is continuing. The present analysis uses damped periodic (Kautz) functions in an attempt to reduce computational errors. Changes to the EDPM program are currently being made to permit the use of additional Kautz pairs to eliminate residual errors. A linear method, based on a pulse analysis in the time domain, has also been developed and is being programmed as a subroutine for computer processing.

Work is continuing on a remote transient method for measuring surface thermal impedance values. This technique employs a high intensity xenon flash lamp to excite a heat wave in the sample surface. The energy absorbed in the sample causes a transient temperature rise at the sample surface. Transformation of this temperature transient yields the surface thermal impedance directly when it is assumed that the driving function is a plane heat wave. Circuitry to accomplish this transformation electronically has been assembled and is undergoing evaluation testing.

Nuclear Ceramics

Outgassing of tungsten-encapsulated UO_2 (during float-zone melting) was eliminated through use of a special grade of tungsten.

A series of sintered UO_2 specimens was thinned to electron transparency by both mechanical and chemical methods.

Preliminary measurements show that the melting point of cerium oxide (CeO_2) is dependent on the atmosphere in the furnace and on the refractory metal used to heat the oxide.

Nuclear Graphite

The microstructural changes produced in graphite during oxidation at very high temperatures have been investigated by electron microscopy. A sample was heated by a spark-machining technique while a jet of oxygen impinged on it in the vicinity of the spark. There was evidence of melting and vaporization of the graphite, and reactions attributed to atomic oxygen were observed.

Graphite irradiation capsule H-3-15, was discharged and is undergoing disassembly.

Graphite irradiation capsule H-3-26, is operating satisfactorily at design temperatures after being discharged in July. The capsule includes 1.7-inch diameter samples of nuclear graphite to check the effect of sample size.

A leak detected in the GEH-13-14 capsule just prior to startup is being repaired. A delay of one cycle will result, and the capsule will begin irradiation in late July.

Approval for the irradiation of an entire subassembly of graphite and thermocouple pins in EBR-II has been requested. The irradiation is scheduled to start in late July and will continue for at least two cycles.

In order to test a recently proposed model for polycrystalline graphite, the density, resistivity, sonic moduli, and CTE of nuclear grade SGBF have been determined and the samples subsequently stressed in tension to various fractions of the nominal tensile strength. A second set of higher density samples is being prepared for the same type of testing.

Fast Reactor Dosimetry and Damage Analyses

To date, surface saturation of fission tracks and irradiation damage have limited the accuracy of mica detectors as fluence monitors for pressure vessel surveillance. Additional irradiations will be conducted to determine if the fluence range of the mica detectors can be extended to higher fluences.

Computer programs are being adapted and checked out which are capable of simulating radiation damage behavior. These behavioral descriptions will assist in the prediction, analysis, and interpretation of radiation damage experiments.

Irradiation Damage to Reactor Metals

Liquid metal capsule GEH-22-5, containing nickel-base alloys, has been sent to the ETR for charging in cycle 91. No change in operation from the previous liquid metal capsule is planned.

A type 304 stainless steel creep specimen prestrained at 450 °C, held at 760 °C without stress, and then creep tested at 650 °C, did not result in improved creep resistance.

Optical metallographic studies of irradiated 410 stainless steel in the annealed condition were completed.

Stress rupture tests on specimens (including control specimens) of Inconel 600 and Inconel X-750 irradiated at 540 °F (282 °C) have been completed. Stress rupture tests on Hastelloy X-280 control specimens representing those irradiated at 1250 °F (677 °C) have been initiated.

A study of the effects of 2500 hours aging at 1250 °F (677 °C) on the subsequent thermal and irradiation stability of Hastelloy X-280 was initiated.

Seven of the 10 participants in the intercalibration program have reported their results to PNL. Analysis of the data received indicates that interlaboratory comparison of Ni and Fe activation results are comparable to data from the 1963 intercalibration program; however, differences of up to 50% in cross sections used by various laboratories have been reported.

Metallography, electron microprobe compositional analysis, and electron microscopic fractography examinations have been made at various locations across the thickness of a 4-inch thick, A302-B, pressure vessel steel plate. These examinations are being conducted to establish the mechanism responsible for the large through-the-thickness fracture toughness gradients present in the plate. To date, the examinations have suggested two controlling mechanisms: (1) the higher toughness near the plate surface is due to smaller colonies of austenite decomposition products than exist at the plate center, and (2) the low toughness at the plate center is the result of the large local variations in manganese concentration giving rise to low toughness in the manganese poor regions.

Specimens of AISI 304 and 348 stainless steels in the annealed and cold worked conditions have been tensile tested after irradiation to 1.7×10^{22} total neutron fluence in the EBR-II. Annealed material showed increases in strength properties at temperatures to 871 °C. The minimum ductility found in annealed 304 and 348 was 10% and 5%, respectively. Cold worked material showed little change in strength, but ductility was reduced by about one-half. The minimum observed ductility for cold worked material was 1%.

The dynamic hydrogen permeation characteristics of A302-B pressure vessel steel have been investigated at 25 °C and 300 °C. The hydrogen desorption behavior does not appear to be diffusion controlled at either temperature.

Installation of the in-cell rectilinear crane in the prototype cell has been completed.

The support system for the sodium loop has arrived on plant. Damage suffered in transit is being evaluated. Preliminary design work has started on the piping for the sodium loop test stands which will be located behind a radiation shield.

The air environment creep test stands are now operating at 1500 °F (815 °C) and 1650 °F (898 °C) with Hastelloy X-280 samples. One unit for inert atmosphere testing has been assembled and is operating at 1832 °F (1000 °C) with Hastelloy X-280 samples.

Repairs, modifications, and the installation of an extension of the horizontal test section of the model loop have been completed. Routine operation of the loop is being resumed.

The adherence and degradation of a superalloy oxide in reducing atmospheres is being considered in relation to the ATR corrosion studies. Thus far, the oxide retention of prefilmed Hastelloy X-280 samples is found to be poor in low hydrogen partial pressure atmospheres as well as in 10^{-4} torr vacuum atmospheres at 1100 °C. The loss of oxide from a superalloy may provide a source of contaminant to the more reactive metals present in the ATR gas loop.

A test to determine the feasibility of using polycrystalline iron and molybdenum specimens in ultrasonic attenuation experiments indicates that the background scattering from grain boundaries is too high to resolve changes in attenuation due to dislocation multiplication during yielding.

Careful high pressure-high temperature tests have revealed that NaK and unfired grade A lava (pyrophyllite) are not compatible above 800 °C and react with a large evolution of heat.

A high pressure form of B_2O_3 with a monoclinic structure has been produced at 39 kbar and 680 °C which is 20% more dense than hexagonal B_2O_3 .

ATR Gas Loop Operation and Maintenance

The design of the ATR gas loop is approximately 90% complete. The critical path item for completion of the loop is the Hastelloy X heat exchanger. Based on current schedules, the stress analysis and design will be completed about September 1, 1967.

PNL is actively engaged in preparing inspections and test procedures for the component parts to determine their compliance with the gas loop specifications.

Metallic Fuels Development

Three Th - 2.5 wt% U - 1.0 wt% Zr fuel elements continue their irradiation in the P-7 hot water loop at the ETR. Exposures range from 9400 to 18,600 MWd/tonne with corresponding volume increases from 1.3 to 3.4%. Dimensional measurements and visual examination of these elements in the TRA hot cell confirm the ETR basin measurements and examinations that are made each reactor cycle. These elements exhibit complete mechanical stability.

Hollow core uranium fuel elements being tested in the ETR-M3 water loop continue to operate successfully in their sixth reactor cycle of irradiation. Estimated maximum exposure at the conclusion of this cycle will be 6500 MWd/tonne.

ENGINEERING DEVELOPMENT

Neutron Flux Monitors

Detailed test planning furthered progress toward comprehensive ETR in-core evaluation of the improved U-234 - U-235 regenerative detectors along with several other types of detectors.

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

A successful reduction of electrical interference noise through a new design of several circuits readied the experimental infrared hygrometer for detailed calibration.

PLUTONIUM UTILIZATION PROGRAM

Fuels Development

Recent fuel-rod examinations associated with the PRTR power calibration have revealed fuel melting at an indicated maximum rod-power generation of 20.0 kW/ft. One of the two rods melted to approximately 10% of the radius; the other rod melted to approximately 33% of the radius. Both rods were presumably irradiated under the same conditions.

One commercially fabricated PRTR fuel rod containing cold pressed and sintered mixed oxide pellets was received from the vendor. Nondestructive examination of the rod was started.

Four fuel rods instrumented to measure fuel-rod gas pressure and temperature continue to operate satisfactorily.

An intentionally defected element containing vibrationally compacted UO₂ fuel was successfully irradiated in the ETR at a maximum linear power generation of 28 to 29 kW/ft for nine effective full-power days.

A redesign of a simple autoclave, which incorporates suggestions made by TREAT Facility personnel, is presently under way. The autoclave will be used for transient testing single fuel rods in the TREAT Facility.

Comparison of high-speed motion pictures of vibrationally compacted UO₂ (5.01 at.% U-235) fuel rods and a vibrationally compacted UO₂ (9.54 at.% U-235) fuel rod undergoing transient irradiation under similar conditions in TREAT showed a difference in failure characteristics.

Nondestructive examination of one vibrationally compacted UO₂ (9.54 at.% U-235) fuel rod subjected to a power transient which resulted in a threshold failure suggests that initial clad penetration is aided by the release of entrained gases in the fuel at elevated temperatures.

Installation of the vacuum welding chamber has been completed except for control wiring.

All equipment for the underwater fuel element handling facility has been received.

A Dresden plutonium-oxide sample was prepared and added to the series of Shippingport PuO₂ samples undergoing systematic gamma dose rate analyses.

Neutron and gamma dose-rate attenuations were measured for some common fuel fabrication containment materials.

Reactor Physics

Batch Core Power Test No. 6, Transient Fission Product Poisoning in PRTR, has been performed, and the results are being analyzed.

Reactor physics assistance has been furnished to PRTR Section to aid in the establishment of operational limits for the batch core in PRTR which will assure no fuel melting.

The contribution of the uncertainties in the value of the cesium-137 half-life and fission yield to the uncertainties of the cross section ratios derived from plutonium-aluminum burnup data has been established.

A calculation has been made of the burnup characteristics for a PRTR loading of irradiated $\text{UO}_2\text{-PuO}_2$ elements driven by Th-U metallic elements.

Design of the core hardware for the joint USAEC-CNEN program in the PRCF is essentially complete, and fabrication of components has begun.

Fabrication of the $\text{UO}_2\text{-4 wt% PuO}_2$ fuel rods has been completed. Initial measurements in the CAF will be in the 0.8-inch hexagonal lattice.

Gamma scanning of fuel rods from EBWR has continued, using the PNL Gamma Scanning Facility.

Gamma ray analysis of Zr-97 - Nb-97 - Mo-97 mixtures has been performed using a Ge(Li) high resolution spectrometer.

A set of equations has been derived which yield the atom densities of selected groups of fission products in reactor fuel as functions of irradiation history, for correlation with gamma scanning measurements.

A summary of a paper entitled "Temperature Coefficients of $\text{PuO}_2\text{-UO}_2\text{-H}_2\text{O}$ Lattices," has been submitted to the American Nuclear Society for presentation of a paper at the November meeting.

A second shipment of fuel irradiated in the EBWR has been made from ANL to PNL.

The power operation of the EBWR Plutonium Demonstration Experiment has been terminated, and postirradiation experiments are being conducted.

Procurement action is in progress to obtain 200 to 600 (4 wt%) PuO₂-UO₂ test fuel rods for use in critical experiments in the PRCF. The two-step procurement procedure is being used to obtain these rods commercially.

Low and high exposure half-inch diameter PuO₂-UO₂ rods were investigated at various square lattice pitches as possible candidates for the initial PCTR water lattice experiments. Preliminary 4-group diffusion theory calculations have been completed to determine the critical number of driver columns for the PCTR and the degree of spectral equilibrium within the test lattice for the case of the water tank assembly loaded with low exposure mixed oxide rods in a 1.00-inch square pitch lattice.

Difficulties encountered with ENDF/B library processing programs continue to delay general use of the system.

Modifications since 1965 to date in the BNW Master Library will be published shortly.

A system of three General Atomic codes designed to generate multigroup constants in the slowing down energy region is being adapted to local use.

Codes to generate scattering kernels for water from the gaussian model now appear to work properly.

Computer codes are being written for use in analysis of gamma-ray spectra and analysis of fission product data from fuel rods.

A new set of ALTHAEA cross section libraries have been generated using the latest plutonium cross section information.

Reactor Engineering Development

Updating of the thermal hydraulics design code, REPP, and corresponding modifications of the survey code, FULCYC, have been completed.

An analysis was performed to determine the adequacy of the fueling vehicle air cooling system to cool the 8-rod FERTF fuel element during charge-discharge. Also, calculations were performed to determine the effectiveness of emergency water cooling systems. It was found that the normal air cooling would be adequate for cooling of a nondefect element. The present emergency water cooling system was found to be adequate to prevent cladding and fuel melting; however, it would probably not prevent rupture of the fuel rods.

Materials Development

Initial examination with the electron microscope of three fractured tubes with varying amounts of hydrogen showed no change in the mechanisms of failure.

Test Reactor Operation

The PRTR operated to 66 MW to produce 539 MWd during July. The reactor power was reduced to 50.5 MW after metallurgical observation of rod from a test fuel element revealed that the previous power was approximately 5% above the desired limit.

The total output from the PRTR is 1901 MWd, of which 1621 MWd have been accumulated by the 55 fuel element batch core.

The Fuel Element Rupture Test Facility (FERTF) was connected to process channel 1550 and returned to irradiation service.

The amount of visible crud on the fuel elements continued to decrease. New 10- and 40-micron pore size filters were installed in the primary system to replace the 5-micron filters. Four process tubes were examined. There were no significant changes observed since the previous examination.

A planned outage for maintenance activities required eight days in addition to 11 unscheduled outages which required 10½ days.

Initial tests were begun for the use of the pneumatic accessory.

EBWR Demonstration Program

The last two capsules in the group of 32 are being examined. These capsules operated with estimated initial linear powers of 14.9-16.7 kW/ft and to estimated burnups of 26,300-27,600 MWd/tonne. Examination of production-run rods, irradiated to an estimated burnup of 27,500 MWd/tonne, is under way.

NUCLEAR SAFETY

Containment Systems Experiment

Calculations of the CSE containment vessel leakage rate were made using each of the individual point temperature sensors, and these values were compared with leakage rates based on the volume weighted average of all temperature sensors. These comparisons indicate that reduced numbers of sensors can under certain circumstances give an adequate sample of containment air temperature.

A second preliminary aerosol transport test was successfully completed in the 2000 ft³ CSE drywell vessel. Preliminary results indicate the half lives in the gas phase was 12 ± 1 min for iodine, cesium, ruthenium, and uranium. Performance of equipment was satisfactory, and techniques were shown to be adequate. This run completed the preliminary testing, and preparations are under way for the next aerosol test which will be made in the 30,000 ft³ CSE containment vessel.

Two blowdown tests were performed on the reactor simulator vessel. In both of these tests, coolant conditions were 500 °F and 665 psig (saturation pressure). The blowdowns were from a bottom nozzle. In one test, a 3.438-inch ID orifice was installed at the end of the nozzle, and in the second test the full bore of the 8-inch nozzle was used (6.8"). The purpose of these tests was to test the adequacy of various mathematical models for predicting the critical discharge of high temperature water following sudden rupture of a reactor cooling system. Transient data on the liquid level, total liquid weight in the vessel, vessel reaction forces, and pressure and temperature in the vessel and discharge pipe were successfully obtained. Preliminary analysis of data indicates that the observed mass flow rate is only about 60% of that predicted by two-phase critical flow models.

The facility used to conduct experiments on the decontamination of Phenoline 302 painted surfaces was modified and calibrated and is now ready to continue experimental operation.

Radioactive Waste Solidification

Pot calcination run PC-4 successfully solidified 650,000 curies of radioactivity in a 12-inch diameter pot to produce 166 Kg of calcine having 2700 watts of self-generating heat and a pot center-line to wall ΔT of 310 °C.

Corrosion rates for 304-L stainless steel in PW-1:NaPO₃ melts at Meq/p ratios from 1.5 to 2.3 were less than 25 mils/mo when exposed at the melting point plus 50 °C. The relatively low corrosion rates indicate feasibility of in-pot melting at these compositions.

Heat evolution rates of 2.6 and 1.1 kilowatts were measured with the in-cell calorimeter for pots from runs PC-4 and PC-2, respectively. These numbers are in good agreement with the values predicted from material balance and decay curve.

Fission Product Aerosol Containment

A spray removal run was completed using 5% hydrazine and a nozzle yielding a mass median droplet size of 140 μ . For equal liquid throughput the removal half-time was only about 1/5 as long

as that calculated assuming that the partition coefficient alone was controlling the removal rate.

An initial analysis of reactive spray removal rates showed that unless fine droplets and/or fairly high concentrations of hydrazine are used, preferably at high throughputs, it is unlikely that 5% hydrazine would double the washout rate in the 10-foot tall spray column.

Reaction rate constants were measured for several reactants with methyl iodide. Rates for hydrazine were similar to those for $\text{Na}_2\text{S}_2\text{O}_3$.

Disposal of Reactor Off-Gas into Soil Systems

Data on the depth to basalt (bedrock) at the proposed NRTS injection site are being evaluated and placed in functional form for use in the flow system analysis program.

Waste Solidification Condensate Treatment

Laboratory studies showed that two-stage electrodialysis treatment followed by anion exchange is an effective method for treating WSEP acid fractionator condensate. A total decontamination factor for ruthenium of greater than 100 was observed. The removal of ruthenium and nitric acid by ion exchange alone was also examined. Results for this method also look promising.

Geophysical Exploration of Rattlesnake No. 1 Well

Results of seven drill-stem tests indicate that communication does not exist to any significant degree between the many basalt flows in the section; and if it does exist, flow will be from the shallower toward the deeper zones due to the higher potentiometric heads in the near-surface flows and interbeds.

Columbia River Sedimentation Studies

Radiochemical analyses of Columbia River estuary samples proceeded on schedule. Retention of several radionuclides on plastic sample containers for reactor effluent water was found to be significant.

Detection of Metal Overstress by Acoustic Emission

Development efforts continued on the program to utilize acoustic emission as a means of detecting flaw growth in reactor pressure

pipng. A system analysis study to consider various parameters and functional approaches for arriving at a practical monitor system is in progress. Three recent burst tests, using 3-inch dia. A-106-B carbon steel pipe samples support two earlier hypothesis concerning acoustic emission. These tests provided good evidence that the continuous background signal which is commonly observed during crack growth is caused by the attendant fracturing of the oxidized surface layer. These tests also indicated that the continuous signal, when present, and also some of the burst or transient signals, are transmitted by surface wave propagation modes whereas most of the other transients appear to be transmitted via shear wave modes.

A laboratory model of a high temperature piezoelectric transducer was assembled and tested for seven hours at temperatures in excess of 600 °F. Transducer performance was encouraging, and satisfactory sensitivity was obtained throughout the test at frequencies up to 500 kHz. The material used for these tests was PZT-5 which is known to have a limited useful life at temperatures above 600 °F. Subsequent models will incorporate piezoelectric materials with higher temperature operating capabilities if such material can be procured.

ASSISTANCE TO CUSTOMERS

The QAD shielding codes have been operational on the local computers. A gamma ray cross section library is being prepared to be used in conjunction with these codes.

Several of the required calculations for fuel irradiation hazards summaries have been or will be transmitted to the interested parties.

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

CIVILIAN POWER REACTORSNuclear Systems and Concepts Analysis (D. E. Deonigi)Systems Analysis Task Force Activities

Checkout runs of modified LP systems, incorporating changes suggested by the Systems Analysis Task Force, have been made throughout the month on the specified AEC Task Force Committee cases (Cases 1 through 7). These changes include: (1) corrected treatment of ThO_2 discharges; (2) narrowed convergence criterion of optimal LP solution; and (3) improved interface with the report writer.

Further modifications to the LP system in progress include: (1) making total fissile plutonium the effective restraint; (2) prorating capital costs of reactor in operation at the end of the problem; and (3) adding a value to the final plutonium stockpile. Cases 1 to 7 will be rerun with these changes incorporated.

Previously suggested modifications to the computer code LPOUT, which generates reports of LP solution, have been made successfully. The modifications in progress include: (1) combining Pu-239 and Pu-241 into fissile plutonium to check the restraint; (2) adding reactor average load factor table to the report; (3) reporting plant capacity and electrical generation comparisons; (4) correcting reprocessing costs to reflect throughput on the same basis as fabrication costs; (5) reporting a reduced cost table; and (6) including actual worth in 1970 dollars of isotope shadow prices as output.

PACTOLUS Modifications

A short program which will simplify methods of handling PACTOLUS and FAST data cards has been written. This code will allow data cards for many cases to be stacked on a tape and executed when a list of desired cases is supplied. These cases will be selected from the data tape, executed, and the output tape to DAEDALUS will be stacked and ready for use in a single pass.

At the same time, FAST is being changed to write to a drum element rather than tape to save the rewind time of a tape. No time comparisons are yet available as debugging is still under way.

In addition, it has been determined that the cash flow PACTOLUS should be referenced to some fixed point in each interval (year) unless the costs are passed to the POWERCO subsection with greater detail as to the times that the various events occur. This will be

done by charging simple interest to the end of each year for events that happen earlier in that year. The programming of this change is under way.

A report (BNWL-429) describing the operation of the PACTOLUS code has been distributed to all Systems Analysis Task Force members.

The PNL version of POWERCO, a subroutine of PACTOLUS, differs from the ORNL version in its treatment of interim capital replacements, and gives a slightly lower calculated power cost for the same yearly percentage of replacements. The PNL program depreciates the interim replacements over the same number of years as the original plant, and writes off the undepreciated portions in the last year. An equivalent amount of original plant investment is written off when replacements are introduced. The ORNL program defines capital replacements differently and treats them as expense items.

POWERCO has also been modified to permit entry of plant salvage value, and the detailed analysis printouts have been changed to provide a more accurate split between fixed and variable costs.

Further modifications are planned to change the treatment of initial core recovery, to permit recovery of D₂O moderator costs and molten salt fuel carrier costs, and to treat property taxes and insurances as part of the plant contribution to power cost.

Offsite Program Requests

An updated FORTRAN II version of the Fuel Cycle Analysis code for the IBM 7090 has been completed, and all links of the code have been stacked on one BCD tape in card image form.

The complete tape will contain four files as follows:

1. A guide to using the tape.
2. The Chain Guide to describe use of the Fuel Cycle Analysis code.
3. The Fuel Cycle Analysis Code.
4. Sample data to be run with the code.

The guide to using the tape, the Chain Guide, and sample data are being prepared and updated for inclusion on the tape.

Uranium Metal Versus Oxide Fueling for Power Reactors

An economic comparison of slightly enriched uranium metal fueling to slightly enriched uranium oxide fueling has shown a fuel cycle cost advantage of about 0.028 mill/kWh_e for metal fuels over oxide fuels. This comparison (see Table I) assumes a fabrication rate of

TABLE I

Effect of Fabrication Rate and Exposure on Fuel Cycle Cost
For Uranium and Uranium Oxide Fuels

Fuel Fabrication Rate, \$/kg	Uranium				UO ₂			
	60	40	20	10	80	60	40	20
Minimum Fuel Cycle Cost, Mills/kWh _e	1.163	1.020	0.862	0.774	1.227	1.127	1.019	0.898
Exposure Required for Mini- mum, MWd/MT	29,010	25,660	21,780	19,600	38,290	35,080	31,200	26,460
Fuel Cycle Cost @ 10,000 MWd/ MT, Mills/kWh _e	1.753	1.426	1.099	0.935	2.144	1.832	1.520	1.207
Fuel Cycle Cost @ 15,000 MWd/ MT, Mills/kWh _e	1.372	1.142	0.912	0.798	1.639	1.424	1.209	0.994
Fuel Cycle Cost @ 20,000 MWd/ MT, Mills/kWh _e	1.227	1.046	0.865		1.418	1.252	1.086	0.920
Fuel Cycle Cost @ 25,000 MWd/ MT, Mills/kWh _e	1.173	1.021			1.309	1.172	1.036	0.899

\$20/kg for the metal and \$60/kg for the oxide and final exposures of 10,000 MWd/MT for the metal and 35,080 MWd/MT for the oxide. Other input parameters are given in Table II.

This analytical result was obtained by substituting uranium metal fuel at 80% TD directly into a typical PWR lattice designed for 90% TD uranium oxide fuel. The rod size used was 0.42 inch OD with a 24-mill clad and a moderator-to-fuel volume ratio of 1.89. A more complete study could potentially arrive at a more optimum core design for metallic fuels.

The calculations were made with the JASON, ALTHAEA, and QUICK links of the Fuel Cycle Analysis burnup chain. Since fabrication rates and attainable fuel exposure are subject to considerable change, the fuel cycle costs have been tabled at several fabrication rates and final fuel exposures in Table I. The assumptions used in the economic analysis and a breakdown of the resultant fuel costs are given in Table II.

As can be seen from Table II, the greatest savings is realized in the increased bred fuel credit obtained from the metal fuel. This is most probably due to the increased epithermal-to-thermal flux ratio which will improve resonance absorption in U-238. Also for the uranium metal case, the fast effect (ϵ) has increased from 1.044 to 1.063.

TABLE IIUranium Metal and Oxide Fuel Cycle Cost Comparison

These results compare fuel cycle costs for a typical PWR fueled with uranium metal or oxide. The uranium is determined from the uranium price schedule, and the plutonium is credited at \$10/g fissile.

	<u>Fuel Type</u>	
	<u>Uranium</u>	<u>UO₂</u>
Plant Capacity, MW _e	1,000	1,000
Exposure, MWd/MT	9,118	35,258
Specific Power, W/cc	221	221
Fabrication Cost, \$/kg	20	60
Reprocessing Cost, \$/kg	20	20
Final Feed Enrichment, wt%	1.20	2.32
Interest Rate, %	12	12
Thermal Efficiency, %	30	30
Load Factor, %	80	80
Density (heavy metal only), g/cc	15.1	8.63
Fuel Cycle Costs, Mills/kWhe		
Depletion Costs	0.6311	0.5775
Losses	0.0381	0.0205
Fabrication	0.3046	0.2363
Reprocessing	0.3297	0.0788
Shipping	0.0990	0.0256
Interest on Working Capital	0.1047	0.1600
Interest on Total Fuel Inventory	0.2607	0.2224
Bred Fuel Credit	-0.6143	-0.2175
Total Fuel Cycle Costs	1.1641	1.1258

Power Reactor Design Studies and Evaluations (J. C. Fox)Steam-Cooled Fast Reactor

Burnup calculations have been run with FCC using ORNL rather than Russian cross sections in an effort to be completely consistent with the rest of the study. Differences in the initial k_{eff} required to obtain a batch burnup of 50,000 MWd/tonne are quite small (0.002).

A set of PNL cross sections is being prepared for use in FCC and comparison with the ORNL and Russian sets.

Current physics results, those presented at ORNL in June, have been added to the design and economics simulations.

Because of the wide difference in out-of-core pressure drop between the B&W Low Pressure Design and the Karlsruhe Intermediate Pressure Design, the simulation has been changed to put out-of-core pressure drop in as a constant value rather than as a fraction of the in-core pressure drop.

The cause of the discrepancy in core pressure drop, as calculated by the PNL design simulation compared to the value given in the D-1 design document, has been found. The difference comes primarily from the way the subassembly flow is orificed. Karlsruhe orificed the flow such that the bulk coolant outlet temperature from each channel is about the same. The PNL simulation assumes orificing such that all subassemblies have the same flow. Since the pumping power must be based on the hot channel pressure drop, the flow in that channel is very critical. However, the increased flow results in better heat transfer, thus a larger flow cross section, and possibly lower pressure drop, for clad temperature limited cores. The orificing design philosophy to use is not completely obvious; therefore, both approaches will be studied using the PNL simulation and the one resulting in the lowest core pressure drop will be used in the parametric studies.

Efforts are continuing on formulating an adequate clad design analysis.

Thermal Sink Limitation Study

Gross heat capacity (i.e., the amount of heat required to raise the temperature of the stream 1 °F) calculations have been completed for the major streams in the South Atlantic Slope Basins, James River to Savannah River and the Eastern Gulf of Mexico Basins, Ogeechee River to Pearl River.

Streams studied in these areas include the Roanoke, Pee Dee, Congaree, Savannah, Altamaha, Swannee, Apalachicola, Choctawhatchee, Alabama, and Pearl Rivers.

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

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

Control, unirradiated tensile tests of Zr-2.5 Nb were conducted for correlation with data reported in the March 1967 monthly report. The results of these tests along with the results of the tensile tests conducted on specimens sustaining prior in-reactor creep are given below:

	<u>Prior In-Reactor Creep</u>		<u>Unirradiated Control</u>	
Test Temperature, °C	350	400	350	400
Ultimate Tensile Stress, 10^3 psi	90.5	70.0	87.7	74.8
Yield Stress, 10^3 psi	63.8	55.1	76.4	70.0
Total Elongation, %	13	21	13	12
Uniform Elongation, %	9	10	6	6
Reduction of Area, %	80	90	81	86

These results suggest that in-reactor exposure under creep stress does not result in strengthening but rather a decrease in strength under the conditions of these tests.

In-Reactor Corrosion of Zirconium Alloys

Coupons of six zirconium alloys were exposed in the ETR G-7 loop for eight ETR cycles, 174 days at temperature. The G-7 loop coolant analysis was pH-10 NH_4OH , <0.05 ppm O_2 . The estimated fluxes are 1×10^{13} and 1×10^{14} n/cm² sec, >1 MeV at the two in-flux positions. Specimens of the six alloys were also exposed in the G-7 loop at two out-of-flux positions with water transport times ~1 sec and ~1 min from the flux. Weight gains at the in-flux and out-of-flux positions appear in Table III.

To summarize the results:

- At the high flux position (Quadrant 234), flux-induced oxidation occurred on all of the six alloys. The oxidation was most pronounced on the Zr-Cu-Fe and prefilmed Zr-Cr-Fe specimens; it was intermediate on Zircaloy-2, Zircaloy-4, and etched Zr-Cr-Fe specimens. The lowest attack occurred on Zr-2.5 Nb and Zr-3Nb-1Sn alloys.
- At the high flux position, the effect of prefilming varied as a function of alloy. On Zr-2.5 Nb and Zr-3Nb-1Sn alloys, weight gains were higher on etched specimens than on pre-filmed specimens. On the remaining alloys, weight gains were higher on prefilmed specimens.
- At the low flux position (Quadrant 233), in-flux and out-of-flux weight gains were comparable for Zircaloy-2, Zircaloy-4, Zr-3Nb-1Sn and Zr-Cu-Fe alloys. In-flux weight gains on the Zr-Cr-Fe alloy were slightly accelerated on etched specimens and greatly accelerated on pre-filmed specimens. Weight gains on Zr-2.5Nb specimens were lower in-flux than out-of-flux, in agreement with the general behavior of this alloy in 2-cycle and 4-cycle exposures.

TABLE III

In-Flux and Out-of-Flux Weight Gains on Zirconium Alloys
After 174 Days in pH-10 NH₄OH

Alloy	Condition	Preautoclave ^a Weight Gain mg/dm ²	Out-of-Reactor BNW ^b Autoclave mg/dm ²	Out-of-Flux		Quadrant 233 1x10 ²⁰ mg/dm ² nv ^c	Quadrant 234 1.7x10 ²¹ mg/dm ² nv ^c
				~1 min mg/dm ²	~1 sec mg/dm ²		
Zircaloy-2	Annealed	E	13.7	-	13.8	15.8	121.0
Zircaloy-2	Annealed	17.8-19.9	-	9 ^d	9.8	8.1	222.8
Zircaloy-2	80% cold work	E	-	-	-	15.0	124.6
Zircaloy-4	Annealed	E	-	-	14.5	14.0	78.8
Zircaloy-4	Annealed	17.2-18.0	-	12 ^d	4.3	9.5	99.5
Zr-1.2Cr-0.08 Fe	"	E	16.3	-	14.4	22.2	117.6
Zr-1.2Cr-0.08 Fe	"	14.0-14.4	-	16 ^d	18.0	393.0	712.8
Zr-1.2Cu-0.28 Fe	"	E	125.1	79.5	44.4	69.3	379.9
Zr-1.2Cu-0.28 Fe	"	19.2-20.5	-	91.8	94.3	105.7	883.0
Zr-3Nb-1Sn	Quenched, Aged	E	-	-	16.2	14.4	44.0
Zr-3Nb-1Sn	" "	20.0-21.9	-	5.2	-	3.3	27.7
Zr-2.5Nb	Quenched, 20% CW, Aged	E	29.0	-	21.8	11.4	52.5
Zr-2.5Nb	" " "	13.4-14.6	-	13 ^d	23.4	3.4	28.1
Zr-2.5Nb	Quenched, 30% CW, Aged	E	30.5	-	17.2	11.6	51.1
Zr-2.5Nb	" " "	12.7-14.0	-	-	14.8	6.6	35.3
Zr-2.5Nb	Annealed, 20% CW	E	18.9	16.0	14.7	12.7	52.4
Zr-2.5Nb	" "	11.9-12.8	-	-	11.8	9.5	41.0

^a Zr-2.5Nb specimens prefilmed 400 °C, 3 days, 100 psi steam; all other alloys prefilmed 400 °C, 3 days, 1500 psi steam. E denotes specimens exposed as-etched.

^b Weight gain at 174 days; autoclaved, pH-10 NH₄OH, 280 °C at Battelle-Northwest.

^c Estimated fluxes; data from flux monitors not yet available.

^d Film thicknesses determined metallographically.

- Fretting attack was obvious on several samples at the 1-min out-of-flux position. Weight gains on unfretted samples and weight gains determined metallographically were in reasonable agreement at the two out-of-flux positions.

The 8-cycle data reported here are the last in a series of experiments conducted in the ETR G-7 loop on the six zirconium alloys. Two-cycle and 4-cycle data from the series appeared in previous monthly reports. In the 2-cycle and 4-cycle tests, in-flux oxidation was only slightly accelerated on etched specimens. In-flux oxidation was pronounced on prefilmed Zr-Cr-Fe, Zircaloy-2, and Zircaloy-4 alloys. The 8-cycle weight gains are generally higher than 2-cycle and 4-cycle extrapolations would predict. The deviation from 2-cycle and 4-cycle data was pronounced for Zr-Cu-Fe, Zr-Cr-Fe, and Zircaloy-2; it was less pronounced for Zircaloy-4; Zr-2.5Nb and Zr-3Nb-1Sn data deviated only slightly from the earlier trends.

The general upturn in in-flux corrosion rate may indicate a radiation-induced transition or, alternatively, a change in the character of the G-7 loop environment. Low corrosion rates at out-of-flux positions, at the low-flux in-pile position, and on certain alloys at the high flux position tend to discount loop contamination as the explanation. An alloy-dependent radiation-induced transition appears to be the more likely explanation. However, the G-7 loop operation, i.e., decontamination, resin changes, and water chemistry, is being examined for unusual factors which might contribute to the accelerated oxidation.

Hydrogen analyses and oxidation data from a 4-cycle prefilm test (Quadrant 243) will appear in the next monthly report.

HTGR Graphite (R. E. Nightingale)

Radiation-Induced Graphite - Water Vapor Reaction

Attempts to measure the rate of the gamma-radiation-induced reaction between TSX graphite and water vapor have continued. Although the water-vapor concentration was raised to approximately 6000 vpm, the oxidation rates in the temperature range 500 °C to 700 °C remain low, e.g., $\sim 10^{-6}$ hr⁻¹. The very small weight changes measured during the reaction were influenced by minor changes in gas flow rate, probably as a consequence of buoyancy effects.

Attempts have been made to determine the reaction rate from the concentration of gaseous products and the gas flow rate. Samples of the effluent gas were taken during runs in which the graphite temperature was as high as about 695 °C, the flow rate about 200 cm³/min, and the water-vapor concentration up to 5800 vpm. Chromatographic

analysis showed no H_2 , CO , or CO_2 in the samples measured, with detection limits of 3, 2, and 4 vpm, respectively. An H_2 concentration of 3 vpm corresponds to an oxidation rate of about $2.2 \times 10^{-6} \text{ hr}^{-1}$ for an 8-gram graphite sample or approximately a factor of from 2 to 10 greater than the rates recently measured by weight changes. It therefore appears that an analytical method of appreciably greater sensitivity will be required to determine the reaction rates by means of the product gases and hence to characterize accurately the reaction of graphite with water vapor as promoted by gamma radiation.

Quantitative Determination of Water Vapor

Many reactions, both thermal and radiation induced, occurring between impurities in the helium coolant in HTGR, as well as reaction of impurities with the graphite, have water as a reactant or a product. It is therefore necessary to have a method for quantitative determination of water at as low a concentration as possible in order to study the effect of impurities on the graphite oxidation.

Methods of analysis based on conversion of water to hydrogen on calcium hydride or to acetylene on calcium carbide followed by standard gas-chromatographic methods of determination of hydrogen or acetylene have been attempted. The calcium hydride used continually gave off hydrogen. Using samples of 10 micrograms of water, the blanks were greater than the samples. This approach was therefore dropped in favor of a method using calcium carbide. Blank experiments average only a small percentage of the total water in our sample. However, recoveries are low, usually about 30% for a water sample of 2 to 60 micrograms, which would necessitate calibration of the carbide columns. The low recovery is believed to be due to holdup of water by calcium hydroxide present as an impurity in the calcium carbide. Attempts are now being made to increase the recovery to the point where quantitative analyses are possible.

Liquid Metal Fast Breeder Reactor (R.E. Nightingale)

Stoichiometry Effects in Oxide Fuels

Twelve UO_2 - 25 wt% PuO_2 fuel specimens with O/M's of 1.92, 1.97, 1.99, and 2.00 were fabricated. Each irradiation capsule contains an $MO_{2.00}$ standard and an MO_{2-x} specimen. Postirradiation measurement of the melt radius in each pin will provide an accurate relative measurement of the effect of stoichiometry on integrated thermal conductivity of the oxide.

Composition gradients which arise in nonstoichiometric oxide fuels during irradiation with no center melting are being studied. Capsules containing $UO_{1.90}$, $UO_{2.00}$, $UO_{2.02}$, $UO_{2.05}$, and $UO_{2.15}$ were irradiated at approximately 20 kw/ft and rapidly discharged

to retain the steady state oxygen distribution. All capsules were returned for metallography and sampling to determine radial composition distributions.

Cooling Rate Effects in Molten Center Oxide Fuels

Variations in structure and fission-fragment distribution in molten center UO_2 fuel elements discharged from the reactor at different controlled rates are being studied. A computer solution of the transient-heat-flow problem was achieved using the TIGER V Program. Results were expressed in terms of the rate of movement of the solid-liquid interface in response to a given power transient. For a rapid scram, the interface moves rapidly initially, then more slowly; and as solidification approaches completion, the rate again increases. For a 1.375-inch diameter fuel core with 60% of the radius molten at steady state, complete solidification occurs 60 seconds after power generation terminates. Other cases including various steady-state heat ratings, fuel sizes, and power transients are being programmed. Results will be used to design in-reactor tests for studying solidification rate effects on fuel structure and impurity distribution.

APPLIED AND REACTOR PHYSICS

Plutonium Criticality Studies (E. D. Clayton)

Studies of Neutron Spectrum

Spectral information is important for interpretation and analysis of criticality data. To date, several spherical proton recoil proportional counters have been obtained in England for use in neutron spectrum measurements over the energy range of approximately 50 keV to 1 MeV. With the proper pulse shape discrimination circuitry it is expected that the useful energy range can be extended down to perhaps 10 keV. The diameter of these small spherical counters is 3.94 cm. The counters contain a very weak plutonium source on the anode wire at the center of the sphere for purposes of calibration.

Most of the work done thus far with the proton recoil counters has been for the purposes of testing, calibration, and measurement of system resolution. The energy resolution for the counters is expected to be about 10% at the energy corresponding to the alpha calibration peak in the one atmosphere detector (359 keV). The resolution of this peak in the calibration tests performed so far has been about 30%, which is greater than desired. In order to more fully examine the problem of energy resolution, nitrogen will be added to the filling gas in two of the counters. This will permit an additional calibration point by means of the $\text{N-14} (n,p) \text{C-14}$ reaction as well as another measure of resolution.

Preliminary measurements of the fast neutron leakage spectrum were performed on an undermoderated plutonium assembly comprised of layers of PuO_2 -polystyrene and Lucite. The base dimensions of the core (a rectangular parallelepiped) were 36.3 cm by 31.0 cm. The estimated critical height for the assembly was 34.9 cm. The assembly was reflected on two sides (ends) by one inch of Lucite. The core contained four 2-inch slabs of PuO_2 polystyrene material (2.2 wt% Pu-240) at an H/Pu of 15, with two inches of Lucite between each fuel layer; the average core H/Pu ratio was 35. Adequate counting statistics were obtained for the spectrum measurements while the assembly was still several dollars subcritical.

The data from these measurements are under study. Computer codes are being prepared for rapid, efficient analyses of the data.

Coincident with the further calibration and testing of these counters, steps are being taken to obtain electronic circuitry for pulse shape discrimination between neutrons and gamma rays to extend the useful range to lower neutron energies.

Mixed Fuel Systems of Pu-238 - Pu-239

The criticality of a mixture of plutonium isotopes containing 18 wt% Pu-238 will be studied in experiments performed with the Remote Split-Table Machine and PuO_2 -polystyrene compacts to provide a homogeneous thermal system. Apart from the application of the data to criticality safety in production of Pu-238, the data will be extremely useful for checking computational methods and cross section sets. As a guide in planning the experiments, a series of calculations were performed to estimate critical dimensions and masses of the plutonium on hand for the experiments. The experiments will be performed with about 5.3 kg plutonium that has the following isotopic content: Pu-238 - 18.2 wt%, Pu-239 - 65.2 wt%, Pu-240 - 10.4 wt%, Pu-241 - 5.4 wt%, Pu-242 - 0.76 wt%. The calculations were made with the HFN code with multigroup constants obtained from the Gamtec-II code. The Pu-238 cross sections were taken from the Battelle master library. An effective PuO_2 density of 6.45 g/cm³ was assumed in conformity with previous PuO_2 -polystyrene fuels. The results of these calculations are presented in the following table:

Criticality of Homogeneous Hydrogen-Moderated
Mixtures Containing Pu-238

Ratio of Hydrogen to Pu (total) Atoms	Bare Spheres		Water Reflected Spheres	
	Critical Radius (cm)	Critical Mass (kg Pu)	Critical Radius (cm)	Critical Mass (kg Pu)
30	24.90	36.40	19.86	18.48
50	25.10	23.33	20.06	11.90
100	24.60	11.33	19.73	5.84
150	24.34	7.40	19.63	3.88
200	24.37	5.60	19.77	2.99
250	24.52	4.58	20.02	2.49
300	24.80	3.96	20.37	2.19
350	25.15	3.55	20.79	2.00
400	25.58	3.27	21.26	1.88
450	26.03	3.06	21.76	1.79
500	26.52	2.92	22.29	1.73

Based on the results of the calculations, taking into consideration the quantity of material on hand, PuO₂-polystyrene blocks will be prepared with an H/Pu ratio of ~400 (~50 g Pu/l). Calculations have been made regarding heat generation from alpha decay of the Pu-238, and it may be necessary to apply some forced air cooling of the assembly core.

Theory and Analysis

The latest version of the British Monte Carlo Code, GEM-3, is now operable on the IBM-7090. Calculations are presently being made to determine the code's applicability and reliability for computing criticality of complex geometries and interacting arrays. A calculation was made for a plexiglas reflected rectangular parallelepiped for which criticality had been determined at the Plutonium Critical Mass Laboratory. The core material was PuO₂-polystyrene with an H/Pu of 15 containing 2.2 wt% Pu-240. The measured critical dimensions were: 23.01 x 23.01 x 20.12 cm. For this system, a k_{eff} of 0.9962 ± 0.0139 was computed by means of GEM-3 code using the UKAEA 7ES184 Pu-239 cross section data.

Containment Tests of Reactor Room

Containment tests required for safety are being conducted to determine the ability of the reactor room to meet original acceptance tests involving internal pressures of up to 4.5 lbs/in². After stopping leaks in the air supply valve, the exhaust stack valve, and in the steam outlet, the leakage rate was materially reduced but is still above the acceptable design limit. Further tests will be conducted on completion of painting and treatment of walls and further sealing around pipes and other inlets.

Phoenix Fuel Reactor Program (D. D. Lanning)

MTR Phoenix Experiment

An irradiation test element has been prepared to test plutonium fuel plates under the operating conditions proposed for the MTR Phoenix experiment. Three series of plates have been prepared. The first plates utilized cast cores which resulted in unacceptable voids and inclusions. The second set of plates utilized extruded cores and had exceptionally uniform plutonium distribution and very few internal voids. These plates used both 6061 and 1100 cladding. The 1100 clad plates utilized a 6061 insert at the core ends to minimize dogboning. Use of the insert resulted in an unbonded interface between the 6061 frame and insert, and attempts to seal the interface by welding were unsuccessful. The third series of plates utilized extruded cores and 6061 cladding only. Again, the plutonium distribution is very uniform and dogboning is essentially nonexistent. Six 9-inch plates and twelve 3-foot plates were prepared and shipped to Idaho Nuclear for hydraulic testing of the assembly. A complete set of external hardware was also fabricated and shipped with the test assembly. Charging of the element will be delayed pending the results of destructive tests being performed on selected plates by BNW and hydraulic tests being performed by Idaho Nuclear.

Preliminary specifications for the Phoenix fuel element and shim rod have been prepared and reviewed with INC and BNW personnel. Revision of these specifications is being prepared for second-step review by all interested personnel. Final specifications will be prepared upon the completion of fabrication and standards development and completion of physics tests.

Use of the 6061 cladding for plates eliminates the use of a brazed shim rod fuel section assembly due to the close melt temperatures of the braze material and 6061.

Fuels Fabrication and Development

Six hundred uranium oxide fuels, 12 cadmium poison follower rods, and 16 follower fuel sections are to be modified for a PRCF fuel experiment. The fuel and follower design work has been completed. Development work for the disassembly, machining, welding, and assembly of components is in process. All new component parts are on order.

PRCF-Phoenix Fuel Experiments

A summary entitled "Critical Experiments with Phoenix Fuel in a MTR Mockup" has been submitted for presentation of a paper at the 1967 Winter Meeting of the American Nuclear Society.

Calibration of one interior and one peripheral shim in the PRCF-Phoenix core has been completed, with the remaining six shims banked at 63.6% withdrawn. The calibration was accomplished by incrementally inserting the interior shim (Shim B) from 63.6% to 38.9% while incrementally withdrawing the peripheral shim (Shim E) from 0% to 100%. Preliminary examination of the differential worth measurements for the peripheral shim yielded a total worth of 49\$. The last 2% withdrawal, from 98% to 100%, exhibited a reversal of worth, equal to -18.1 cents. This phenomenon is believed to be the result of moving the lower end of the fuel follower out of the high thermal neutron flux just below the core, while the upper part of the fuel follower was being moved into the region at the top of the core where the flux is severely depressed by the banked shims. The total worth of the interior shim appears to be about twice that of the peripheral shim, or about 18\$.

Borated polyethylene sheeting will be used to simulate equilibrium xenon poisoning in the core. Measurements of the reactivity worth of a single sheet of the plastic (23.5 x 2.5 x 0.005 inches) attached to the center plate in the center fuel element, again pointed up the sensitivity of this core to small disturbances to the fuel element positions. Successive measurements, each of which require removal and reinsertion of the center fuel element, produces values of -20¢, -13.7¢, and -8.2¢, respectively. Thus, it appears that reactivity measurements which require removal and replacement of fuel elements will have inherent uncertainties of the order of 5-10¢. This problem will receive further study.

PRCF-Phoenix Fuel Calculations

A survey of the experiments completed indicated the desirability of performing at least one additional experimental loading such that one or more symmetrically-placed control rods can be fully inserted with the fuel followers removed. The goal of such an experiment would be to determine the critical size of a configuration involving control rods which would be amenable to a two-dimensional calculation. The ability to calculate in two dimensions allows the use of more than two energy groups, whereas our three dimensional code is currently limited to only two energy groups. A strong group structure dependence was found for the critical approach experiments, and it is expected that a similar effect will be found for the PRCF series. Hence, the differences in k_{eff} due to collapsing to two groups must be known on cores with control rods if comparisons of three dimensional, two-group calculations are to be meaningful. The indicated experiment is currently planned for the last set of PRCF experiments.

In order to best satisfy the requirements for the diffusion theory approximation, a homogeneous model for the core was selected. To compute thermal spectrum averaged cross sections for the entire core, including the aluminum side plates and the thin water regions between fuel boxes, two related THERMOS calculations were performed.

First, spatial fluxes were computed over the meat, clad and coolant slab unit cell. Next, another slab geometry calculation was performed for the water between boxes, the side plate, a smeared region of water and aluminum representing the alternating aluminum plate edges and coolant channels, and, finally, the active region. In the last region the entire meat-clad coolant unit cell was homogenized, but the nuclei densities were adjusted according to the region-averaged flux ratios obtained from the first case. The resulting averaged cross sections are then correct for the entire core unit cell, including the structure and coolant located beyond the fuel plate ends. Indeed, the spectrum is sufficiently softened by the inclusion of these regions so that the microscopic absorption cross section of Pu-239, for example, is increased by 5%.

MTR-Phoenix Fuel Calculations

Investigation into the large difference in the magnitude of the power peaks between the PRCF experiment and the MTR 3-D calculations has continued. Because the thermal group parameters for the fuel have the greatest effect on the power peaking, an alternate source for these parameters was sought.

The original MTR calculations were performed using thermal group parameters from the HAMMER system version of the THERMOS code. This code was used because it calculates a cell diffusion coefficient in addition to the normal cell output parameters. The alternate source chosen for the thermal parameters was the BNW modification to the THERMOS code.

The BNW version of the THERMOS code calculates a cell diffusion coefficient in a different manner than the HAMMER version of the THERMOS. For unit cells consisting of thin slabs, as is the case for the MTR fuel plates, the thermal diffusion coefficient from THERMOS/BNW is approximately 2.5 times as large as the diffusion coefficient from THERMOS/HAMMER. The difference in the other cell parameters from the two codes was 2% or less.

Using the thermal parameters from THERMOS/BNW in a 3-D calculation simulating the PRCF control rod position, but with MTR fuel compositions, it was found that k increased approximately 0.5 mk while the peak-to-average power ratio ($P/\bar{P}$) decreased from 3.07 to 2.70.

Comparisons were then made between the measured and newly calculated power distributions in selected fuel plates. These comparisons exhibited the same 10% decrease in the power peaks at the bottom water reflector-fuel interface as was found in the $P/\bar{P}$ ratio. It was found also that the power peak below the core in the shim fuel was traced more accurately both in magnitude and shape.

Thus, it appears that more investigation into the calculation of the thermal diffusion coefficient is in order. The actual value of the coefficient seems to have little effect on the eigenvalue but have large effect on flux shapes next to moderating regions.

Code Conversion. A compiler/optimizer error was found in THERMOS/HAMMER whenever stacked cases were run in which the number of mesh points was varied from case to case. The error resulted in the number of mesh points in the last case of the stacked set being used in all cases in the set as the index limit in the calculation, the total macroscopic cross section, and the total source in the cell.

This error has no effect if the last case of the stacked set happens to have the largest number of mesh intervals. However, if the last case has fewer mesh intervals than all other cases, very small negative values for the neutron density will occur at each mesh interval greater than the number of intervals in the last case.

Changes have been made to the THERMOS programs to alleviate this condition, and the program tape is ready for use.

A document entitled "WHIRLPOWER/POWERWRITER - Two Group, Three Dimensional Codes to Calculate Normalized Flux and Power Distributions" is in final publication process.

MTR Core - Thermal Hydraulic Analysis

Present consideration sets the MTR operating power for a Phoenix-fueled core at 29 MW_t. The thermal and hydraulic limits used in the analysis were based on past operating experience of the reactor and are more stringent than authorized limits. A pressure head of 60 psia and a pressure drop through the core of 40 psi were maintained in the investigation. The maximum allowable values are 75 psig and 50 psi pressure loss. Since boiling burnout and incipient nucleate boiling are strong functions of the system pressure, the effect of pressure head variation on reactor power is being investigated. Preliminary estimates indicate a maximum operating power of 40 MW_t may be attainable.

In the first set of preliminary specifications, the fueled section in the shim rod follower was designed for 12 fueled plates with 0.126 inch coolant gaps. In an effort to increase fuel quantity and heat transfer surface area in the core, a preliminary analysis was conducted to determine the feasibility of increasing the number of plates from 12 to 13. This could be accomplished by using coolant gaps, consistent with the stationary fuel element, of 0.119 inch. It was found that this allowed 0.065 inch coolant gap each for the two outside coolant channels. Although the coolant in the outside channel is subjected to heat flux from only one heated surface, the 0.065 inch channel gap did not meet heat transfer

requirements. However, increasing the gap to 0.085 inch thickness provided the necessary heat transport conditions. This requires removing 0.020 inch from the end plate reducing the thickness of the end plate from 0.237 inch to 0.217 inch.

MTR Core - Mechanical Design

A different flux monitor wand is being considered which consists of a solid aluminum 0.115 inch diameter rod with a 0.020 inch diameter Pu fuel core. The original design consisted of a fuel cored 0.050 inch diameter wire within an aluminum tube, with coolant flow in the annulus between the wire and tube. The solid wand will be easier to fabricate and less susceptible to damage during handling. Calculations show that adequate cooling of the solid wand will be provided by the coolant flow along the side of the wand that is open to the flow channel between the fuel plates. The wand is inserted down beside the fuel side plate between the second and third fuel plate from the concave side of the fuel box.

MTR-Phoenix Experiment

An irradiation test element has been prepared to test plutonium fuel plates under the operating conditions proposed for the MTR-Phoenix experiment. Three series of plates have been prepared. The first plates utilized cast cores which resulted in unacceptable voids and inclusions. The second set of plates utilized extruded cores and had exceptionally uniform plutonium distribution and very few internal voids. These plates used both 6061 and 1100 cladding. The 1100 clad plates utilized a 6061 insert at the core ends to minimize dogboning. The use of the insert resulted in an unbonded interface between the 6061 frame and insert, and attempts to seal the interface by welding were unsuccessful. The third series of plates utilized extruded cores and 6061 cladding only. Again, the plutonium distribution is very uniform and dogboning essentially is nonexistent. Six 9-inch plates and twelve 3-foot plates were prepared and shipped to Idaho Nuclear for hydraulic testing of the assembly. A complete set of external hardware was also fabricated and shipped with the test assembly. Charging of the element will be delayed pending the results of destructive tests being performed on selected plates by BNW and hydraulic tests being performed by Idaho Nuclear.

Preliminary specifications for the Phoenix fuel element and shim rod have been prepared and reviewed with INC and BNW personnel. Revision of these specifications is being prepared for second step review by all interested personnel. Final specifications will be prepared upon the completion of fabrication and standards development and completion of physics tests.

The use of 6061 cladding for plates eliminates the use of a brazed shim rod fuel section assembly due to the close melt temperatures of the braze material and 6061.

Applications

CNSG Studies. Physics information on Phoenix fueled loadings of various pressurized water reactor designs has been calculated and transmitted to personnel conducting economic evaluations of these loadings. The designs are prototypical of the Consolidated Nuclear Steam Generator. A rough draft of a report describing the physics calculations and results is being written.

Changes were made to REPP program to supply the option of holding the mass flow rate constant when searching for a heat-transfer-limited reactor core.

Heat-transfer-limited cores for fuel volume fractions from 0.40 to 0.50 and pin diameters of 0.25, 0.35, and 0.43 inch have been calculated. The initial enrichment requirements and end-of-life inventories are being found for reactors covering the above range. Fuel cycle costs will then be determined.

Cores with geometries similar to the original CNSG-II core have also been analyzed. One of these is to be used as the base case and has UO_2 and poison pins, the other has PuO_2 and no poison pins.

HFIR. Data sheets, both input and output, for the ORNL methods used to calculate HFIR systems have been received. Work is now under way to carry out similar calculations using PNL methods in order to check the applicability of our methods to compact flux trap reactors.

High Temperature Reactor Lattice Physics Studies

(R. E. Heineman)

Reactor Construction

At the end of June all major construction on the High Temperature Lattice Test Reactor was complete. Design testing was under way on the heating and gas cooling system. The project was, therefore, officially closed out with the exceptions to be completed with accruals to the project. The reactor heatup test was terminated on July 2, because of a failure in an electrical control circuit. The reactor temperature was 675 °C. The reactor was then cooled to room temperature for an inspection of all components and to prepare for nuclear design tests.

The drive assemblies on three vertical safety rods were modified to match the fourth unit which was tested during the heatup design tests. These modifications have been completed and are ready for final adjustments. The vertical safety rod housings were modified to accept the "rod-in" limit detector. During the course of this work one drive and its housing (but not the rod) fell from the top of the reactor to the floor. There was some minor damage to the

housing and the inner liner and to the alignment of the drive itself. The liner was replaced; the housing and the drive unit were repaired. A chain hoist being used to support the rod during modification of the housing was shown to be at fault.

The reactor operations personnel installed the active portions of the control rods. Inverse multiplication measurements were taken during this work since the rods contain enriched uranium fuel. The control rod drive units were mounted on the reactor shell and coupled to the rods. Shim collars were fabricated for the control rod drives. These act as a positive stop to avoid damage of the diaphragm in the event of over-run of the system and limits. The rods were being checked out at the end of the month.

The two oscillators were brought to the reactor building at the conclusion of the heatup and cooldown tests. The layout work for attaching them to the reactor shell was completed. Wiring for the power and control circuits was essentially complete.

After the horizontal control rods were installed, an interference was found between the rod drives and the rear hatch. A modification to the hatch was completed.

Two leaks were located on the reactor shell at two of the VSR penetrations. Repair was rather difficult because the welding was almost inaccessible. Leak checks will be made following the installation of the vertical rods.

Reactor Equipment

Oscillators. All work on both oscillators was completed, and the oscillators were delivered to the reactor building for installation whenever space in the reactor cell is available.

Horizontal Control Rods. During the installation of the active portions of the control rods, several outer graphite sleeves were broken. The break occurred at a thread relief point, and the installation procedures were modified to accommodate this weak point. The break could be expected to occur only during transfer of the rod and not during normal operation.

Vertical Safety Rods. Installation and testing of one gas jet-type in-limit switch was completed satisfactorily using the prototype housing in 314 Building. Installation procedures were prepared together with the jet units. Installation was completed on three of the rods.

While work was being done on VSR #1, a chain hoist supporting the rod assembly failed, and the unit fell from the top of the reactor to the floor. Significant damage was done to the rod housing and rod channel extension. Some damage was also done to

the stepping motor and drive. The complete unit was returned for disassembly, examination, and repair. The upper housing was straightened, and the top flange was repaired; the rod guide channel was badly damaged and was replaced; the stepping motor was removed and returned to the factory for repair; and the brake release cylinder was damaged and was replaced with the one from the prototype assembly.

Programmed Measurement and Control System (PMACS). Some contact outputs in PMACS are failing, apparently because of under-rating of contact capacity. Arc suppression circuits were recommended to eliminate this problem. A failure of the ADC was also noted during this report period. Intermittent failure of a diode in the conversion circuit was causing the problem, and it was replaced.

The flux monitoring equipment in PMACS was checked in preparation for nuclear tests. However, a failure of the flux digitizers, which convert μ p amp currents from the ion chambers to digital information, terminated the testing. The flux digitizers were returned to the vendor for repair. One has been returned, and the other is ready for shipment.

An ammeter has been installed to read current through the main gas blower with a readout to PMACS. Similarly, a vibration monitor has been installed as recommended by the manufacturer.

Reactor Operations

The high temperature heating and cooling test run was terminated on Sunday, July 2, due to loss of control on the heaters. The reactor temperature was 675 °C at that time. An inspection revealed that diodes and fuses had been burned out in the electrical control circuits. The loss probably resulted from high ambient temperatures (140 °F) in the outside substation, located in full sun. These control circuits have been removed and installed in the basement.

The reactor was cooled and the doors removed for inspection of all internal components on July 9. Several graphite bolts from the rear bus bars were observed to have broken. Subsequent inspection shows that bolts on the front bus bars had also broken. Inspection and laboratory tests showed that the bolts had failed in tension by high stress. They had been inadvertently fabricated with the grain orientation opposite to that of the bus bars. Replacement bolts were fabricated and installed.

Some Kaowool and other dust was noted in the fuel channels during the inspection following the test. The remainder of the graphite, insulation, and doors appeared in excellent condition. Leak tests were made and significant leaks were found in two of the VSR wells. These have been rewelded.

Following the heat run, all holes in the graphite with marginal dimensions were honed with a special tool designed for the purpose. All graphite sleeves and plugs were inserted, as well as the horizontal control rods.

As a result of the test run, minor modifications were made to the controller on the process water pumps, to the piping on the gas dryer, and to the flange on an expansion bellows on one of the bus bars.

A relay coordination study for the HTLTR primary power system was also completed. The relays and circuit breakers have been set and tested at the 318 Building and substation C3S4 in such a way that the building service is not interrupted in the event that the breakers on the heater power system open.

A review of the programs used in the tests, GAS CYCLE, HEAT, COOL, and various LOG programs, was completed. Work has been done in five areas: (1) Debugging gas and heat system programs, (2) Implementing changes to the gas-heat system programs, (3) Writing a valve calibration program to be used in conjunction with the gas-heat programs, (4) Writing the ADC calibration routine, and (5) Developing sampled data techniques.

Debugging Gas and Heat System Program. The trouble with logging the four average temperatures, which was reported last month, was cured. During several days of operation, no trouble was traced to this fault.

A mistake in the fixed point automatic control calculation in the gas system programs was found and eliminated. The trouble showed up as an intermittent failure which caused certain valves under automatic control to fly open or closed. An add and clamp routine was shortened to gain more core area. The shortened routine failed to account for all the possibilities of sign and overflow with the "ADD" instruction.

Another bug affecting automatic control was found and eliminated. After a little less than four hours running time, the valves would not go into automatic control upon request. This had never shown up before because all the tests were for less than four hours. This was eliminated by allowing 17 bits for elapsed time rather than the full 18 bits.

A bug that hasn't been eliminated seems to be affecting only one group of valves. The valves designated 1-, 2-, and 3-TCV12 will not go into automatic control.

Changes in the Gas-Heat System Programs. A few changes in the gas-heat programs were initiated because long term operation showed them to be necessary. There are three changes in the gas system and

three in the heating system programs.

Operator control of the protected process water pumps via the keyboard will be eliminated. The room is needed for other, more important, functions. Also, there is a hazard that the operator will not turn a pump on through the teletype keyboard after the system program has stopped and has been started again. The operator will have to start and stop a selected pump manually.

Presently, the gas system program automatically switches to the emergency cool state if the analog position feedback for any digital valve fails to read properly. This will be changed so that the operator gets an error message.

Before considerable operating experience with the gas system was obtained, it seemed that valve 1PCV12 could be automatically controlled with feedback from pressure transducer 1PT12. This has proved to be untrue. A section of the gas system program would change 1PCV12's setpoint to reduce gas flow to the reactor when the cooling water from the main heat exchanger became too hot. When a method is found which will decrease cooling gas flow when the main heat exchanger is too hot, it will be incorporated in the program.

The heating system is not as well balanced as was initially expected. For this reason, maximum heater limits, which can be changed individually through the teletype, will be put on each heater bank. Presently there is one limit for all four heat banks.

To gain room for the above change, the rundown routine will be removed from the heat system program. The rundown routine stepped the heater control signal down to zero in 0.25% (of full scale) steps. This will be changed so that the control signal is set to zero. The heater power will not change abruptly because of the capacitance in the long lines from the heat DAC's to the heater controllers. Since the power does not drop to zero when the control signal does, a timer will be added so that the heater breaker will not close until the control signal is zero.

Valve Calibration Routine. Because of the play in the mechanical linkage that positions the valves, an auxiliary valve calibration routine is needed. Design of the routine has been started. This routine may be requested to calibrate one valve. When the calibration routine is called, the gas system programs must be active.

ADC Calibration Routine. The ADC calibration routine in the original form has been debugged. Changes to calibrate the high precision RTD channels at all nine scale ranges were introduced into the program. These changes have not been debugged yet.

Sampled Data Techniques. A computer program was written in Fortran, to run on the PDP-7, which will invert modified (or ordinary) Z-transforms. Because the program so greatly simplifies what has previously been a difficult task, the technique has been written in a form suitable for submission to the Proceedings of the IEEE as a letter.

Reactor Physics

Startup and Calibration Experiments. The loading of the horizontal control rods has been completed. Even with all of the HCRs in and open, the reactor is so subcritical and the fuel so spread out that no accurate statements may be made. However, from the approach curves it appears that the reactivity change from open-to-closed (the bank worth) is approximately equal to loading four HCRs in their open positions, as would be expected.

Counts taken while raising a vertical rod indicates that one VSR is approximately equal to the worth of the 8 HCRs operated as a bank.

While setting up for this test, it was discovered that no counts could be obtained using the BF₃ provided with the oral monitor. This is due to the high filling pressure of the tube. The high pressure produces pulses which are too small to drive a long cable. A new chamber, with a lower filling pressure, is being obtained to replace the original BF₃.

Design of the experimental oscillator couplings, front and rear support tables, and associated equipment was completed. Fabrication of the support tables was started. Design of the oscillator carrier strips and support buckets was started.

U²³³O₂-ThO₂ Experiments. In order to better duplicate the type of lattice proposed for high temperature graphite-gas-cooled reactors, the initial U²³³-ThO₂ experiments have been revised. The lattices will have the fuel in small diameter fuel columns with low density rather than the densely packed 1" rods which were first to be used in the HTLTR. The pitch of the fuel holes will be about 7/8".

Agreement was reached with Oak Ridge National Laboratory to convert the U²³³O₂ (mixed 1 part U-233 with 3 parts Th) into microspheres and coat with pyrolytic graphite. This involves the construction of a U-233 facility which will take about three months. Delivery of the coated particles should begin around December 1, and experiments using them could start around January 1, 1968.

Due to the high cost, it was decided not to produce ThO₂ microspheres but to use fused ThO₂ instead. This type of ThO₂ would consist of irregular particles of close to theoretical density that

could be screened to give particle sizes within known tolerances. The larger $U^{233}O_2$ - ThO_2 particles will be able to be separated from the ThO_2 by screening.

Contact with General Atomic on these proposed experiments was maintained, and a GA review of the lattice, experiments to be done, and schedule is in progress.

As previously reported, the Nordheim scheme for evaluating resonance integrals has been incorporated into the GAMTEC II code. The new code GAMNIT has been debugged and is presently in operation evaluating $U^{233}O_2$ - ThO_2 lattice arrays that are of interest to the HTLTR experimental program. With the addition of the Nordheim integration scheme, GAMNIT has the capability of generating multi-group constants reflecting the improved treatments of resonance capture for the broad and lower energy resonances.

REACTOR FUELS AND MATERIALS

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

Synthesis of Mixed Nitrides by a Carbothermic Reduction Method

Kinetic measurements on nitride synthesis via the carbothermic reduction of the oxide were made for UO_2 and UO_2 - 20 wt% PuO_2 . The reaction under study is:

The reactions were carried out under a constant flow of nitrogen, and the rate was followed by monitoring and recording the fraction of carbon monoxide in the exhaust gas.

Mixed nitride syntheses were carried out in a 4" x 3/4" molybdenum boat lined with tungsten foil. The solid reaction mixture was prepared by ball milling 80 g UO_2 , 20 g PuO_2 , and 10 g graphite for 24 hours, pressing the powder into pellets, and crushing the pellets into a -60, +100 mesh powder. Ten-gram portions of stock material were then reacted at a set temperature in a graphite furnace to obtain the kinetic data.

To date, kinetic measurements have been taken at five temperatures between 1430 and 1625 °C. The reaction appears to be pseudo 1st order and to vary with temperature in a nearly Arrhenius manner. The measured rate constant at 1430 °C is $6.1 \times 10^{-3}/\text{min}$ and at 1625 °C is $2.5 \times 10^{-2}/\text{min}$. The calculated Arrhenius activation energy is 6.2 kcal, and the calculated reaction half lives are 113 minutes at 1420 °C and 31 minutes at 1625 °C.

Rate studies for the synthesis of uranium nitride were carried out in 1" OD x 4" alumina crucibles in a molybdenum-wound furnace. Micronized UO_2 was milled with 9 wt% graphite of 1-5 micron particle size to make the reaction mixture; a 4-5 gram synthesis batch was used.

Rate measurements were made at five temperatures, but the data haven't been completely analyzed. However, one rate measurement at 1420 °C gave a first-order constant of $1.9 \times 10^{-3}/\text{min}$.

Kinetic measurements will be completed as soon as possible, and comparison of uranium nitride and mixed nitride reaction rates will be made. The kinetic effect of physical properties will also be investigated if rate data for the two nitrides differ widely.

UN-PuN Synthesis*

Experimental work was begun during the month using the 8-inch spray calciner to produce UO_2 -carbon mixtures starting with UNH-sugar solutions. Five experimental runs were made using feed solutions of varying sugar-to-uranium ratios. In general, the uranium and HNO_3 content was held essentially constant at 1.51M and 1.5M, respectively, while the sugar concentration was steadily increased from 240 to 420 g/l. In four of the runs the calciner vibrator was not operated during the run or subsequent cooldown. The powder in the collection pot was sampled and removed. Then the calciner was vibrated, the filters were blown back, and the recovered powder was sampled. In one run the calciner was vibrated continuously during the entire run to minimize powder holdup on the hot walls. Quantitative analytical results are not yet available on the uranium and carbon content in the powders. However, some results are available on preliminary ignition studies where the powders are held at 950 °C in air for 2.5 to 4 hours and the weight loss determined. These initial results and visual observations are summarized as follows:

1. At sugar concentrations of 240 and 320 g/l, very fine uniform powders were produced and ignition tests indicate about 5 and 8.5 wt% carbon, respectively. These carbon contents are expected to be slightly low since a black residue of carbon remains after ignition and subsequent dissolution of the residue in HNO_3 .
2. At sugar concentrations of 360 and 420 g/l, the powders consisted of a heterogeneous mixture of fine black powder and brownish agglomerates roughly 1/16 to 1/4 inch in diameter.

*Although related to the Fast Reactor Oxides and Nitrides Program, this study is supported by the AEC Special Studies Account.

3. About 10 to 25% of the powder is held up on the calciner walls and filters when the filter blowback system and vibrator are not operated during a run. Ignition tests indicate that this powder contains about 0.5 to 1% carbon. Thus, frequent filter blowback and vibration will be required to minimize carbon burnout.

Irradiation of Mixed Nitrides

Two experimental fuel capsules (GEH-14-745 & 746) containing mixed nitrides (UN-20 wt% PuN) continue to operate satisfactorily in the MTR. The capsules have accumulated a maximum exposure of approximately 21,000 MWd/tonne while operating at maximum heat generation rates that have ranged from ~25 to ~32 kW/ft. These capsules are scheduled to be discharged from the reactor on July 31.

Two previously irradiated capsules (GEH-14-743 & 744) were returned to Hanford for postirradiation examination. The capsules accumulated exposures of ~2000 and 5000 MWd/tonne, respectively.

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 P-10 was discharged after operating successfully at design conditions of 450 °C and 5000 psi. The capsule contains specimens of several dilute uranium base alloys and high purity uranium. The status of capsules currently active is indicated below.

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

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

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

High Pressure Postirradiation Annealing

The second postirradiation high pressure-high temperature annealing study has been completed. The results of the test are listed below.

Density Measurements of Samples Subjected to Postirradiation Annealing at 15,000 psi, 900 °C for 101 Hours

Sample No.	Sample Description	Burnup %	Density at 20 °C		R = $\frac{\% \Delta V / V_0}{\% \text{ burnup}}$	
			Before ⁽¹⁾	After	Before ⁽¹⁾	After
QC2	U-U diffusion couple	Control	16.52	16.55	Not applicable	
QA2	↓	0.46	16.42	16.31	"	
QB2		0.46	16.56	16.51	"	
QC12		0.46	16.99	16.87	"	
PC2	½" dia. x	Control	18.92	18.95	-	-
PA2	¼" long	0.37	18.77	18.55	3	6
PB2	cylinders	0.37	18.81	18.43	2	8
PlA14-2	Same as above but vacuum annealed at 600 °C for ½ hr prior to pressure test	0.37	17.72	18.62	19	5
T12	Split tube	0.15 ⁽²⁾	18.15	18.38	32 ⁽³⁾	23
T22	0.030" thick	0.15	16.87	17.69	85 ⁽⁴⁾	50

(1) Before pressure test but after irradiation.

(2) Irradiated at 575 °C, 1000 psi.

(3) High purity-high carbon (400-500 ppm).

(4) High purity-low carbon (10 ppm).

From the table it can be seen that the application of pressure at high temperature has actually densified the samples which have previously swelled due to irradiation at high temperature and considerably retarded the swelling of samples irradiated at low temperature. Optical metallography and electron microscopy is currently under way to study the nature of the aligned tearing which was present in the tubular specimens before the test and also the bubble size distribution in the cylindrical specimens. Of special importance will be the comparison of the bubbles present in sample PlA14-2 to

those in PA2 and PB2 because of the similar R values obtained from such different annealing histories.

The U-U diffusion couples are also being photographed in anticipation of a high temperature anneal at one atmosphere. This anneal will hopefully reveal the position of the fission product gases so that measurements of diffusion coefficients can be made.

Nondestructive Testing (J. C. Spanner)

Eddy Current Test Methods

Basic Studies. A study has been started to determine the feasibility of using an automated system to adjust the parameter separating circuits of the multiparameter eddy current tester. Test circuits for applying the multiparameter test techniques to a pulsed eddy current system are being assembled, and a detailed analysis is being made of the functioning and adjustment of the parameter separating circuits of the multichannel eddy current tubing tester. The effect of test coil replacement and instrument drift on the reproducibility of instrument output is receiving special emphasis.

A project has been undertaken to determine the feasibility of replacing the present manual method of separation of parameters in the multiparameter eddy current test by an automated method.

The present manual method of separating four parameters requires handsetting the values of 16 coefficients (potentiometers) in the matrix equation $(P) = (a) (c)$; where the signals, (c) , are operated upon by the coefficient set, (a) , to produce parametric outputs, (P) . With this method, large amounts of time are required to optimize the settings for the four parameter cases.

The physical problem has been arbitrarily split into two segments; an automatic coefficient analyzer and an interface from the analyzer to the test apparatus.

With regard to the analyzer section, automatic calculation of a truly linear case is felt to be a reasonably straightforward problem from a device standpoint. However, the parameter vectors described are merely linear approximations to the true locus of variation. This requires an optimization of results on some criterion such as "signal-to-noise" or least-squared value of amplitude of undesired signals. Work is focusing upon selecting the best means for performing this optimization.

The problem of interfacing equipment cannot receive a great deal of attention until a firm selection of the type of equipment

for the analyzer section has been made. If the automatic method appears feasible, a prototype system will be designed and constructed for evaluation.

The multiparameter pulsed eddy current laboratory test arrangement now consists of a pulse generator, two R-C simulating networks, a differential signal amplifier, an orthogonal exponential signal analyzer comprising orthogonal exponential generators and signal sampling unit, and, finally, a signal summing network. Circuits have been made operational up to the exponential signal generators and sampling unit. Circuits for alternate arrangements to evaluate different pulse analyzing techniques are also being provided.

Reproducibility of flaw information and a definite calibration procedure are of primary concern at this stage of development of the multichannel eddy current tubing tester. The parameter separating circuits used in this tester are being used because they can be easily and accurately adjusted for a certain set of parameters providing these are all the variables that are allowed to change. An analysis is being performed, treating the detected signals out of the 100 kHz and 300 kHz channels of the tester as being proportional to the product of trigonometric functions of angles and the magnitude of the associated parameter vector. The tester has two pair of outputs. One pair provides quadrature signals that are derived from a 100 kHz driving frequency, and the other pair provides quadrature signals from the 300 kHz frequency. The analysis performed thus far, indicates that reproducible results can be obtained providing the relative phase angle between the probe motion vectors from the two pair of outputs is maintained constant. Also, it appears that signal to residue ratios could be improved if it were possible to vary this phase angle. Possibly the best setting would be with the phase angle of the probe motion vector at 45° for each of the two pair of outputs with respect to a given arbitrary coordinate axis. If further analysis and empirical data prove this to be true, a phase shifter may be included for each pair of outputs from the next model of this tester.

Ultrasonic Test Methods

Ultrasonic, mechanical, and electronic apparatus has been assembled, and steel specimens are being irradiated for the purpose of developing a test method to detect irradiation induced shift in the nil-ductility-transition temperature in structural steels.

The various electronic units have been integrated into a single package, and a linear RF gate chassis was developed. An RF gate and a peak amplitude pulse stretcher circuit are now being developed to obtain a varying DC output whose amplitude is proportional to the peak amplitude of the central zone of the received pulse. The central zone is the part that varies as a function of the irradiation exposure. Modifications to the air-lock cell in 324 Building are being made to enable operation of the test system within the cell.

Wave Propagation Studies. Analysis of ultrasonic wave propagation in a media with attenuation continues. In previous reports the problems associated with predicting the response of attenuated broad frequency band pulses were discussed. Briefly, it had been found that round-off errors were the likely causes for the poor representation of pulses by Fourier Series approximations which employ uniform periodic functions. A new analysis using damped periodic, or Kautz, functions was developed in an attempt to reduce computational errors.

The problem of replacing the uniform amplitude functions with Kautz functions has been, and is currently, in two distinct parts. First, since the given ultrasonic data are in the form of amplitude values at discrete time intervals, they must be fitted to Kautz functions; and secondly, the ultrasonic response pulses after propagation through the attenuating medium must be computed from the response frequency spectra.

In studying the first problem, the method which had appeared to be best for fitting the ultrasonic data to the first pair Kautz functions used only the first two data points in estimating the Kautz function amplitudes and phase. This method is acceptable so long as the ultrasonic data are by nature derived from a pair of Kautz functions. However, subsequent estimates using as many as four Kautz pairs to eliminate residuals have not successfully represented the ultrasonic data.

In successfully estimating the pulse frequencies and damping factors, a method which includes all the given data points was used. It is now expected that inclusion of all the available data should also better estimate the amplitudes and phases of Kautz pairs. This change is currently being made in the EDPM program.

As mentioned above, the second part of the problem involves reconstruction of an attenuated pulse from its spectrum. This computation was presumed to be nonlinear. However, a linear method, based on the previously discussed method for pulse determination in the time domain, has been developed. The EDPM subroutine is currently being debugged, and tests to assure the validity of the numerical process will be conducted.

Thermal Test Methods

Work is continuing on a remote transient method for determining surface thermal impedance values. The method utilizes a high intensity xenon flash lamp to excite a plane heat wave in the sample surface. The energy absorbed in the sample due to the incoming light wave causes a transient temperature rise at the sample surface. The Laplace transformation of the temperature transient yields the surface thermal impedance of the sample directly, with the assumption of a plane heat wave into the sample.

A laboratory electronic system to evaluate the Laplace transform automatically has been constructed as reported previously. Difficulties with the Barnes radiometer used to detect the temperature transient were encountered, and the instrument was subsequently sent to the factory for repairs. Preliminary tests on the repaired instrument indicate that it is functioning satisfactorily; more extensive tests will be needed to determine this conclusively.

While the Barnes radiometer was undergoing repairs, construction of a simple radiometer was undertaken in the laboratory. The detector used was a commercially supplied indium antimonide variety which required liquid nitrogen cooling. The detector exhibited sufficient sensitivity to detect the sample temperature transient. Preliminary tests on the system indicated that it would meet the requirements of the intended application. However, the dewar flask containing the detector element has started leaking so the flask will not hold liquid nitrogen. The infrared window is constantly covered with condensate because of the loss of vacuum in the flask. Hence, the detector cannot be used without being repaired. Extremely poor quality construction has been encountered in all the commercial detectors recently acquired.

Composite Test Methods

Ultrasonic-Eddy Current. Two types of systems for use in compensation of the effect of transducer characteristics on ultrasonic nondestructive test signals have been studied. The first system using an operational amplifier with an adjustable feedback circuit is least effective because some adjustments were excluded due to circuit instability. The feed-forward technique, the second system evaluated, uses several operational amplifiers. Circuit techniques are being sought by which the number of amplifiers used can be reduced.

Two high frequency operational amplifiers have been procured having unity gain-bandwidth equal to 100 MHz for use in this investigation.

Nuclear Ceramics (R. E. Nightingale)

Growth of PuO₂ Single Crystals

Tungsten-encapsulated UO₂ was successfully zone melted without the outgassing problem experienced previously. The capsule for this experiment was formed from nuclear-grade, chemically vapor-deposited tungsten tubing which had been precleaned and examined microscopically for freedom from surface cracks. Although beam power was maintained throughout the zone pass at a constant level, melting occurred only at the free end of the capsule. Since the power required for melting increases with position along the axis from the free end,

direct control of temperature is necessary for a stable molten zone. The system is currently being modified to allow direct optical pyrometer measurements of sample surface temperature.

Thinning of Nuclear Ceramics for Transmission Electron Microscopy

A series of sintered UO_2 specimens was thinned to electron transparency by both mechanical and chemical methods. Preliminary examination by transmission electron microscopy indicates that even under closely controlled conditions, surface damage is introduced during the mechanical thinning steps. Attempts will be made to remove this worked surface by a brief chemical etch. This should result in larger and more evenly thinned specimens than can be prepared by chemical thinning alone.

Two pneumatically-impacted specimens were prepared from the same ceramic grade UO_2 powder as the sintered pellets. These will also be thinned by both mechanical and chemical techniques to compare sintered structures with pneumatically-impacted structures.

Melting Points of Cerium Oxide

Preliminary measurements show that the melting point of cerium oxide (CeO_2) is dependent on the atmosphere in the furnace and on the refractory metal used to heat the oxide. Initial melting was observed between 1900-2000 $^{\circ}\text{C}$. Repeated melting of the same sample increased the melting point to a maximum value which depended on the atmosphere and refractory metal heater. The results are summarized below.

Maximum Melting Point ($^{\circ}\text{C}$) of CeO_2

<u>Heater Material</u>	<u>Atmosphere</u>	
	<u>8% H_2-Argon</u>	<u>Argon</u>
W	2380	2160
Rh	2365	2330
Ta	--	2090

Some reactions between the refractory metals and the CeO_2 were observed.

Nuclear Graphite (R. E. Nightingale)Gas-Graphite Reactions

Work is continuing on a study of the microstructural changes produced in TSX and AGOT-LS graphites by reaction with oxygen under various conditions. During the past month investigation of the effects of high temperature thermal oxidation reactions was initiated. This part of the study is conducted using a modified spark-machining technique in which the oxidation takes place within the regions immediately surrounding the sparks. The temperatures which have been produced in graphite in these regions appear to extend at least to the melting point under certain conditions.

In principle, a spark machine operates by maintaining a series of sparks between a tool and the item to be machined. An electrical potential of a few hundred volts is applied across the spark gap, and the position of the tool is adjusted continuously, by a servo mechanism, to maintain a constant series of sparks. Spark size is adjustable to achieve coarse or fine machining as desired. The environment of oil which normally surrounds the spark gap was removed for the present series of experiments. Instead, the spark gap was positioned a few millimeters from the orifice of a small oxygen jet, flowing at 200 cm³/min. The tool was a 1/8-inch diameter spectrographic graphite rod which had been sharpened to a fine point. To date, two experiments have been performed with TSX graphite. The first was an attempt to machine a hole through the thin central area of a disc specimen, and in the second the hole already present in a standard mechanically-thinned disc specimen was further machined. The first experiment was not successful because the point of the spectroscopic graphite tool was dulled too rapidly to be effective. It appears that this type of graphite is chemically or mechanically removed considerably more rapidly than TSX graphite under the circumstances of the experiment.

In the second experiment large changes were produced in the graphite microstructure, even with the smallest spark size available, in 1-min oxidation treatments. Two significant observations were made after this experiment. First, there was considerable deposition from the spectroscopic graphite tool on the TSX specimen. Second, for both the TSX and the deposited spectroscopic graphite, there was appreciable lace structure development of a type similar to that developed in graphite oxidized either in or downstream from the glow of microwave-excited oxygen. Since oxygen is known to decompose into oxygen atoms to a significant degree at the temperatures believed to exist during these experiments, and since ozone is unstable at such temperatures, it appears that the lace structures were produced by atomic oxygen. Such structures have not been observed in specimens oxidized at low (600-800 °C) temperatures where molecular oxygen is the only gaseous reactant.

Irradiation of "Proof Test" Graphites

Graphite irradiation capsule, H-3-26, was charged on July 12, and taken to temperature after the GETR startup on July 14. All nine Geminol thermocouples are operable, and all 13 dials are at design temperatures. The capsule is scheduled for discharge in November.

This capsule contains four large samples (1.7" dia. by 2.25" long) to determine the effect of sample size. The grades represented are: CSF, NC8, 67-3, and TSGBF. The design of these larger dials is compatible with the modified capsule series to be initiated in November 1967. The modified capsules will permit the study of sample size and external restraint on the rate of radiation-induced dimensional changes.

High Temperature Irradiations

After the GEH-13-14 capsule had been charged in the ETR for cycle 90 startup, a pressure leak was detected when the capsule portion of the lead-in tubing was pressurized to 250 psi. The capsule was removed from the reactor, and a leak was found in a thermocouple extension lead. The leak is being repaired, and the capsule is now scheduled to be charged during the cycle 91 shutdown in late July. The irradiation will be terminated in December after three cycles of exposure.

EBR-II Irradiations

Prospects appear favorable for irradiation of an entire subassembly in a Row 6 position in EBR-II. Maximum flux in this position is about 1×10^{15} nv (E = 0.18 MeV). The irradiation will begin in late July and will be probably run for two cycles. The determining factor for discharge is the need for space for higher priority capsules.

All pins required to fill the subassembly have been shipped to EBR-II. These consist of 16 graphite capsules, two thermocouple capsules, and one dosimetry capsule. The graphite irradiations include samples that were previously irradiated to exposures of about 10^{22} nvt in the EBR-II. The current irradiation will allow us to continue testing the dimensional stability of some sample types that appeared promising at 10^{22} nvt. An annealing series, previously irradiated in the ETR, will be included in the experiment. A variety of new graphite types is also included.

Results from the thermocouple irradiations are expected to assist in the selection of a reliable thermocouple type for use in fast reactor systems. The dosimetry pin will test flux and exposure determination techniques in EBR-II in addition to providing fluence values for the subassembly.

Investigation of Graphite Structure

A simplified model of the macrostructure of polycrystalline graphite has been proposed by O. D. Slagle (see BNWL-SA-1237). The model allows the calculation of certain properties of polycrystalline graphites in terms of the corresponding properties of single crystals, given the spatial distribution function for crystallite c-axes. Since the model is intimately involved with the microcrack system of the material, any change in that system should show up in the measured properties in a predictable way. Such a change can be introduced by stressing samples in tension to various levels. A program has been undertaken using parallel samples of SFBF nuclear graphite to be stressed at 0, 30, 60, and 90% of the nominal breaking stress. The properties being measured are apparent density, electric resistivity, flexural and shear moduli (sonic), and coefficient of thermal expansion to 400 °C. Average values of these properties for the 16 samples in this group are shown below.

<u>Property</u>	<u>Temp. °C</u>	<u>Average Value</u>	<u>Total Spread, %</u>
Apparent density	25	1.607 g/cc	1.6
Electrical resistivity	25	$8.46 \times 10^{-4} \Omega\text{cm}$	6.8
Flexural modulus	25	$1.283 \times 10^6 \text{ psi}$	8.7
Shear modulus	25	$0.2605 \times 10^6 \text{ psi}$	8.0
Coefficient of thermal expansion	25-425	$2.27 \times 10^{-6}/^\circ\text{C}$	11.0

All of the samples have been stressed and are currently being prepared for remeasurement. A second set of samples of high density graphite ($\sim 1.9 \text{ g/cm}^3$) is being readied for the same type of testing.

Fast Reactor Dosimetry and Damage Analyses

(R. E. Nightingale)

Pressure Vessel Dosimetry*

Studies are being conducted to determine the feasibility of using fission-track detectors as fluence monitors for reactor pressure vessel surveillance. To date, surface saturation of fission tracks and radiation damage have impaired the accuracy of mica detectors as fluence monitors. A gradient in track density was noted as successive layers were cleaved from the samples. Prior to irradiation the samples of mica were wrapped in aluminum foil to facilitate handling, and it is possible that aluminum knock-on atoms contributed to the track production near the surface of the mica. It has been reported that only atoms with masses greater than or equal to silicon can produce tracks in mica. However, since aluminum

*This study, although related to fast reactor dosimetry, is funded under a Special Studies account.

is close to silicon in weight, aluminum knock-on atoms may have caused tracks. It is not clear whether the track formation is a function of the atomic mass of the incident particle only, or is dependent on the ionization potential or a combination of each. Based on the ionization potential, it might be concluded that a metallic knock-on atom (e.g., Al) would be more completely ionized than a nonmetallic atom (e.g., Si). This ionized metal atom would deposit energy in the mica through ionization interactions causing tracks to be formed in a manner similar to that of a fission fragment. Irradiations will be made to provide an indication of the causes of track formation in mica. This information will permit the selection of a cover material which will not contribute additional tracks or cause radiation damage to the mica.

Mica samples wrapped with aluminum and Mylar were irradiated and are now being analyzed. Initial results indicate that the track density in the aluminum-wrapped mica is much greater than that covered with Mylar.

Samples of mica are being prepared for irradiation in the Materials Testing Reactor. The purpose of this irradiation is to determine if the fluence range of mica detectors can be extended beyond 5×10^{18} nvt ($E > 1$ MeV) by using cover materials other than aluminum. Cover materials of beryllium, mica, and polyimide plastic coated with Teflon will be used to reduce radiation damage and track formation by serving as catchers for atoms that could produce tracks.

Adaption and Operational Checkout of Computer Programs for the Computer Simulation of Defect Behavior

Several computer programs are being adapted which are capable of calculating lattice-atom configuration, stability, and movement. They were designed to describe the generation of primary knock-on atoms, the configuration of the collision cascade, the damage state which results, and annealing processes for these defects.

The SPECTRUM Program calculates the spatial and energy spectral distribution of the primary knock-on atoms (PKA) produced by an incident neutron spectrum. The CASCADE Program utilizes the PKA spectrum to compute the interaction with the atomic lattice structure. This gives a description of the disordered region, which may be a simple cluster or defect for a low energy PKA, or it may be an extensive defect spike. The DEFECT Program is particularly useful for calculating the configuration energy or relative stability of a given defect, the energy required or produced when the defect is moved to another position, and its preferred or most stable configuration. The ANNEAL Program utilizes the information produced by the DEFECT and CASCADE Programs to describe what happens to the disordered regions as they attempt to find a stable configuration.

These programs are in various stages of adaption and checkout. Several of the early calculations are oriented towards particular problems which have been selected for their current interest and application. The first is the study of the vacancy-migration energy near a void, and the lattice configurations or criteria associated with the growth and shrinking of the void. A second problem, which is also amenable to calculation, is the study of the configuration energy and the interstitial migration energy of helium near a void. The third problem is the calculation of fast-neutron-produced defect clusters and spikes, and their classification by size and complexity, for ultimate comparison with the reported high temperature displacement effects on embrittlement of stainless steel.

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

Alloy Selection

The emittance of gas from the pressure tubing of liquid metal capsule GEH-22-4 has been traced to a pinhole defect through the weld metal joining the pressure tube with the capsule body. The defect was sufficiently small to survive all quality control tests and 55 days of reactor operation before evidence of gas leakage forced discharge of the experiment.

GEH-22-5 containing nickel-base alloys Hastelloy X-280, Inconel 625, Inconel 718, In-120, and Incoloy 800, has been completed and shipped to NRTS. The irradiation is planned to follow the pattern of previous liquid metal capsule irradiations. At full reactor power, the specimens will be exposed at 1100-1400 °F (593-760 °C) to sodium containing 18 ppm oxygen and 25 ppm carbon to a planned fluence of 1×10^{21} n/cm² (E > 1 MeV).

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. Current effort involves measuring creep in AISI 304 stainless steel, nickel-base alloys, and refractory metal alloys during neutron irradiation.

There is evidence that in some of the 300 series austenitic stainless steels a substructure developed at a high stress will result in improved creep resistance at lower stresses and higher test temperatures. Neutron irradiation during the development of the substructure might provide further stabilization of the substructure and be an effective method of strengthening. The in-reactor creep capsule is well suited for this type of study, since the substructure during irradiation can be developed under a closely controlled temperature and stress, and the strain can be closely monitored. In a preliminary study, an unirradiated specimen of

annealed 304 stainless steel was pre-crept to 18% strain under a stress of 50,000 psi at 450 °C, held at 760 °C for 137 hrs without stress, and subsequently tested at 650 °C under a stress of 20,000 psi. The resulting strain rate was $1.0 \times 10^{-3} \text{ hr}^{-1}$. The corresponding strain rate in a specimen given 18% strain at 650 °C and 20,000 psi was $9.9 \times 10^{-4} \text{ hr}^{-1}$. No significant difference in the creep resistance representing the two pretreatments is apparent.

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.

Optical metallography on annealed 410 stainless steel irradiated to a fluence of $6.44 \times 10^{21} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) (quadrant 80) is complete. There are no detectable structural changes observable with the optical microscope that would account for the low ductility (2% uniform) at the 500 °C test temperature. There were no microcracks observed in any of the specimens. An abstract covering irradiation effects to annealed 410 stainless steel was submitted to the American Nuclear Society for presentation at the November 1967 meeting.

A study of the effect of a long-time age at 1250 °F (676 °C), followed by irradiation at the same temperature, on the postirradiation tensile properties of various thermal/mechanical treated stainless steels is being conducted. Specimens are presently being aged at 1250 °F (676 °C) to a goal exposure of 2500 hrs. The aged specimens will be divided into three groups: the first group will be tested without further treatment; the second group will be irradiated at 1250 °F (676 °C); and the third group will be given control thermal treatments.

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

A paper, entitled "The Influence of Thermomechanical Treatments on the Strength, High Temperature Stability, and Microstructure of Hastelloy X-280," will be presented at the International Conference on the Strength of Metals and Alloys to be held in Tokyo, Japan, in September 1967.

A paper, entitled "Thermomechanical Treatments of Hastelloy X-280 for Nuclear Reactor Applications," has been accepted for presentation at the Fall Meeting of the American Institute of Mining, Metallurgical, and Petroleum Engineers in Cleveland, Ohio, in October 1967.

The stress-rupture testing program on Inconel 600 and Inconel X-750 specimens irradiated at 540 °F (282 °C) and on corresponding control specimens has been completed. (The tests on similarly irradiated Hastelloy X-280 specimens, and their controls, were completed earlier.) Data from these tests are being analyzed.

Stress-rupture tests have been initiated on unirradiated Hastelloy X-280 specimens that are controls for those irradiated at 1250 °F (676 °C).

A study of the effects of long-term aging on subsequent thermal and irradiation stability of Hastelloy X-280 was initiated during the last month. Standard and experimentally treated specimens were placed in an aging furnace at 1250 °F (676 °C) for a goal exposure of 2500 hrs. Half of these specimens will be irradiated at 1250 °F (676 °C) to a goal fluence of 1×10^{20} n/cm²; the other half will be given control exposures at 1250 °F (676 °C) for an equivalent length of time. This work developed from an experiment to determine the effects of irradiation at 1250 °F (676 °C) on the mechanical properties of Hastelloy X-280, Inconel 600, and Inconel X-750. The mechanical properties revealed significant thermal instability of standard treated material. This instability was found to be reduced by experimental treatments which caused significant precipitation to occur prior to 1250 °F (676 °C) exposure. These treatments also produced a substructure that was more stable to neutron irradiation. Long-term preirradiation aging treatments are being conducted to determine if preirradiation stabilization of the microstructure can result in even better postirradiation properties.

Intercalibration Test of Counting Laboratories. To date, seven of the ten participating laboratories have reported activation monitor counting results to the Pacific Northwest Laboratory. Each laboratory received two sets of Fe, Ni, Ti, and AlCo (0.1% Co) monitors which were irradiated in the MTR VH-2 Rabbit Facility for two hours and eight days, respectively. Each set was counted before shipment to the participants.

A tabulation of the standard deviation of count data from eight laboratories for each type of monitor is presented in Table IV. These results indicate that differences in counting activated monitors are not large enough to cause appreciable error in the comparison of radiation damage data. The standard deviations calculated from these results are only slightly greater than those from the second intercalibration test held in 1963, despite personnel changes in many of the laboratories and new supplies of monitor wire.

TABLE IV

INTERSITE COMPARISON ON FLUX MONITOR

Reaction Counted	Percent Standard Deviation			
	Specific Activity		Total Activity	
	2 hr Irr.	8 day Irr.	2 hr Irr.	8 day Irr.
^{60}Co	4.18	4.41	3.66	4.14
^{58}Co	6.43	6.74	6.08	6.81
^{54}Mn	3.48	5.63	3.76	6.06
^{46}Sc	5.61	1.98	5.54	2.11

Although the counting data would indicate that no real problem exists in comparing fluence from irradiation tests, fluxes calibrated from these data definitely show otherwise. Each laboratory was asked to calculate neutron flux and fluence from their count data, given the appropriate irradiation history, and the neutron spectrum for the irradiation facility. The results received so far show that cross sections used by several of the participating laboratories for the $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and the $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ differ by as much as 50%. These differences, which are carried through the reduction of data to the fluences reported, are significant and cause unacceptable uncertainty in comparison of radiation effects data.

Fracture Mechanics

The results of fracture toughness testing of a 4-inch thick A302-B pressure vessel steel plate have been discussed in previous reports. In summary, it was found that there exists a very strong gradation in the plane strain fracture toughness, K_{IC} , across the thickness of the plate, the toughness being lowest in the center and highest at the plate surface. The compressive yield strength is also lowest in the center of the plate; however, the thickness gradation in yield strength is considerably smaller than toughness. One generally finds that the toughness is inversely related to the yield strength. The direct relationship between fracture toughness and yield strength, as was found in the A302-B plate, suggests that the mechanisms controlling the fracturing process are more strongly dependent on the microstructural character of the material than are macroscopic plastic flow properties. While the strong, through-the-thickness toughness variations pose a major obstacle to determining the effect of specimen thickness, per se, on toughness (the original intent of this study), the variations do offer an opportunity for correlating the fracturing behavior with microstructure or other properties of the steel.

A series of examinations across the A302-B plate thickness have been initiated to provide an explanation for the fracture toughness gradient. These examinations have so far consisted of metallography, electron microprobe analysis of certain constituents, and electron microscopic fractography.

From the metallographic examinations it was found that, throughout the plate thickness, the microstructure consists essentially of a fine dispersion of carbides in a ferrite matrix. The fine carbides were spheroidized by the relatively high temperature (1200 °F, 650 °C) of the tempering treatment given the plate. Several differences in the microstructure between the plate center and surface were noted. The over-all carbide-ferrite morphology was fairly uniform near the plate surfaces, but a banded structure existed throughout the remainder of the plate. These bands, which are spaced approximately 100 microns apart, possess an overabundance

of carbides while the regions between the bands are somewhat denuded of carbides. The former austenite grain boundaries are also more clearly delineated near the plate center than at the surfaces.

Electron microprobe scans to ascertain the extent of local variations of the manganese, molybdenum, and carbon concentration have been performed at the surfaces and center of the plate. The average concentrations of these three elements were found to be about the same in both the center and surface of the plate. However, the distribution of these elements was much more uniform near the surface than in the center. High concentrations of manganese and molybdenum were identified with the bands in the plate center. The carbon concentration was also considerably higher within the bands which suggests that the manganese and molybdenum are primarily in the form of carbides. The regions between the bands are poor in manganese and molybdenum.

Fractography performed on the fractured DCB specimens indicates that the primary mode of fracture is cleavage throughout the plate thickness. However, some distinct differences in the cleavage morphology were observed between the center and surface of the plate. The cleavage facets on the broken specimens from the plate center are relatively large and readily identifiable. However, the fracture surface of specimens from the surface of the plate was characterized by small, rather indistinct, cleavage facets surrounded by regions of ductile tearing.

These various examinations have provided several important clues concerning the underlying cause(s) of the through-the-thickness fracture toughness gradients. Of the many possible mechanisms controlling the fracturing behavior, two are presently being considered. One of these mechanisms is based on the colony size of austenite decomposition products. It is known that plastic deformation causes a reduction of the colony size which in turn raises the fracture strength by decreasing the extent of unrestricted cleavage crack propagation. Regions near the surface underwent more plastic flow than the center of the plate during hot rolling to a 4-inch plate. An alternative mechanism being considered is based upon the effect of manganese on the intrinsic surface energy of low carbon steel and the corresponding energy required for initiating a running crack. While the electron microprobe results indicate that the average manganese concentration is uniform throughout the plate, the manganese denuded areas between the bands present in the center region of the plate may possess a lower surface energy. The latter mechanism infers that an improvement in the homogeneity of the steel plate in terms of the alloying content would have a beneficial effect. It is also probable that a weld zone which is a location of compositional heterogeneity may possess regions of low fracture toughness because of the lack of certain surface energy enhancing elements like manganese. Efforts to establish the controlling mechanisms are continuing.

Fast Reactor Supporting Studies

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

Tensile testing of AISI 304 and 348 stainless steel specimens irradiated to a maximum total dose of 1.7×10^{22} n/cm² in the EBR-II has been completed. Both annealed and 25% cold worked conditions were investigated, and effects of specimen orientation with respect to rolling direction were studied for 348 stainless steel. The irradiation temperature has not been accurately calculated as yet, but preliminary estimates suggest values in excess of 600 °C.

The irradiation produced a marked increase in 0.2% offset yield strength in annealed 304 and 348 steels. The increase in strength was more than two times the pre-irradiation value for test temperatures between 25 and 600 °C. Above 600 °C, the strength drops rapidly and approaches the control value at 750 °C in the 304 but persists to 871 °C in the 348. The increase in ultimate strength was not as great as the change in yield but followed the same general trends in temperature.

The minimum uniform ductility in the annealed 304 stainless steel was about 10% at 750 °C. This value is much greater than found in earlier experiments on a different heat of material in which a minimum of 0.5% was observed. The irradiation temperature is also believed to have been somewhat lower in the earlier experiment than for the present work. The differences in ductility loss between the two experiments will be analyzed further to establish the nature of the difference in response to irradiation between the two heats.

Uniform elongation in annealed 348 in specimens cut from both the transverse and rolling directions was decreased by about a factor of two by exposure in the EBR-II. Minimum uniform elongation was about 15% below 650 °C, decreasing to 5% above 700 °C.

All cold worked material showed little change in strength properties after irradiation. Uniform elongation was, in general, somewhat improved below 600 °C. At higher temperature the ductility decreases by about a factor of two. The minimum ductility found in any of the cold worked material after irradiation was about 1%.

Fast Neutron Mechanisms. Fundamental studies of hydrogen behavior in reactor metals are continuing, using both hydrogen permeation and gas evolution techniques. The dynamic hydrogen

permeation characteristics of quenched and tempered A302-B pressure vessel steel have been investigated at 300 °C and room temperature using, respectively, gaseous and electrolytic hydrogen charging methods.

At 300 °C, the hydrogen absorption behavior is diffusion controlled with a hydrogen diffusivity of 1.1×10^{-4} cm²/sec. The corresponding desorption behavior, however, is controlled by an as yet undetermined surface or internal interaction mechanism and follows a rate law of the form $1/J$ proportional to t^2 , where J is the desorption flux and t is time.

At room temperature both absorption and desorption behavior are anomalous. Desorption follows the rate law $1/J = a + B \cdot t$ and is unaffected by the nature (aqueous, vacuum) of the desorption environment. Further work is in progress to clarify the mechanisms involved for both temperature ranges.

Thin Section Materials. The program to select a material for an in-core pressure device for the FFTF is continuing. It was anticipated that tensile specimens from thin sheet material would be exposed to flowing sodium in the 314 Building isothermal loop and then tested in tension. Sheet specimens in the thickness range from 0.005 to 0.015 inch have been prepared. Preliminary results from specimens exposed in the isothermal loop indicate that adjustments in compatibility must be made to serve the needs of the program.

Test Facilities. The bid due date for the liquid metal storage and loading facility, which will include an inert atmosphere glove-box, was extended to the last week of July at the request of the vendors.

Design for the renovation of Building 321-A is in the comment stage. A date for the start of construction has yet to be set.

The MSAR support system for the sodium loop was received on plant during the month. The sodium loop support system, when off-loaded from the railroad car, showed evidence of severe shock during transit. Supports were broken or knocked out of position and pipe insulation was cracked, broken, and displaced. Damage is being assessed, and a damage report is being written.

Preliminary design work has been started on the piping for the sodium loop test stands which will be located behind a radiation shield. This effort involves detailed consideration of required mechanical functions, thermal stress analyses, and radiation safeguards requirements.

Currently, creep tests in the conventional test stands are being performed on Hastelloy X-280 at 1500 °F (815 °C) and at 1650 °F (898 °C) in air. One bench mounted test stand for an inert atmosphere

environment has been assembled and placed in service. Hastelloy X-280 is being tested in helium at 1832 °F (1000 °C).

ATR Gas Loop Supporting Studies

Model Gas Loop Studies. The model gas loop is a 2100 °F (1150 °C) recirculating helium loop which is being operated to determine the effect of helium atmosphere on high temperature 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 3935 hrs since installation of the new heater with 75 thermal cycles from room temperature to various heater outlet temperatures. No operating time was accumulated during the past month; however, repairs and modifications to the system and preparations for a new test series are nearing completion.

An addition to the horizontal test assembly which will permit insertion of the test specimens into the hot gas stream after the loop is at operating conditions with high purity helium is completed and currently being evaluated. This system is being developed in order to conduct corrosion tests under extremely well-controlled gas conditions. The eventual goal is to allow insertion, testing, withdrawal, and inspection of specimens without shutting down the loop system.

Tests with flowing gas at room temperature and 1000 °F (538 °C) have been conducted. No increase in the gas impurities was found during the room temperature test; however, at 1000 °F (538 °C) a small increase in the hydrogen impurity level was noted. No increases in the impurity levels of O₂ - CO - CH₄ - N or H₂O vapor were detected. These tests were conducted with the bypass cleanup system shut down after clean gas had been obtained. Similar tests with the cleanup system operating are being conducted.

Superalloy and Refractory Metal Corrosion Studies. The adherence of oxide films on metals is of continuing interest in the ATR corrosion program. The structural metals put into a high-temperature inert-gas loop will most probably be lightly oxidized. This oxide is therefore a source of contamination to the more reactive metals planned for loop exposure, notably niobium and niobium alloys. The presence of oxide suggests potential problems resulting from (1) mechanical failure of an oxide film, freeing particulate matter to the gas stream and (2) chemical reduction of the oxide, freeing oxidants at low partial pressures to the helium stream.

A laboratory-scale experiment was started to investigate more fully structural metal oxide mechanics. Essentially, the procedure is as follows:

- a. Oxidation of a Hastelloy X-280 sample at 1000 °C in pure oxygen to about 1 mg/cm² sample weight gain,
- b. Holding the sample at temperature while the oxidant is pumped out,
- c. Subjecting an oxidized sample to either 10⁻⁴ torr dynamic vacuum or 0.02 torr replenished hydrogen at 1100 °C, or
- d. Subjecting an oxidized sample to either 10⁻⁴ torr dynamic vacuum or 0.02 torr replenished hydrogen at 1100 °C, and
- e. Continuously following the weight change of the sample.

The oxide formed at 1000 °C and held for about two days at 1000 °C in 10⁻⁴ torr dynamic vacuum seemingly retained its protective nature. However, spalling of the oxide did occur when the sample was cooled to room temperature.

Oxides formed at 1000 °C and exposed to 10⁻⁴ torr dynamic vacuum for two days at 1100 °C immediately lost continuity, and weight losses resulting from metal evaporation, oxide spalling, oxide reduction, or a combination of the three were noted. The same behavior was also observed for a sample oxidized at 1000 °C and exposed to flowing low pressure hydrogen at 1100 °C. The isothermal weight loss in both tests followed essentially a smooth nonlinear curve, indicating no gross oxide spalling from the samples. Weight loss amounted to 1.12 mg/cm² during a 26-hr period for the sample exposed to vacuum and 1.33 mg/cm² during a 25-hr period for the sample exposed to hydrogen. After each sample had cooled to room temperature, oxide degradation occurred to the extent that large oxide flakes could easily be wiped from the sample surface.

The combination of high temperature and a reducing or inert atmosphere plays an important role in superalloy oxide degradation. Further work to separate those mechanisms promoting oxide loss by chemical or mechanical means which are typical of ATR conditions is planned. Also, testing an oxidized superalloy in near or intimate contact with a reactive metal in a reducing atmosphere is planned.

Damage Mechanisms in Iron

The objective of this program is to determine how defects produced in iron by neutron irradiation interact with moving dislocations to modify the mechanical properties of the metal.

During this period a paper describing the experimental techniques employed in the study of thermally activated flow in Ferrovac E iron and the theoretical significance of the results as applied to bcc metals in general was completed for submission to Acta Metallurgica.

A test to determine the feasibility of employing polycrystalline specimens for ultrasonic attenuation experiments was performed. The results indicated such specimens are unsatisfactory due to the large scattering from grain boundaries. In the case of iron the first echo could not be distinguished from the background while for a molybdenum polycrystal the first echo could be distinguished, but changes due to dislocation multiplication during yielding could not be resolved. Previous tests on Mo single crystals had permitted resolution of four echos and a change in attenuation with strain had been observed.

High Pressure Studies

The previously reported formation at 20 kbars and 900 °C of an unknown alloy or compound of Mo, Ta, NaK, pyrophyllite and 303 stainless steel has been repeated under carefully controlled conditions. In the most recent test the temperature was slowly increased until the reaction took place. This reaction is accompanied by a large volume change of the reaction products. Previously determined power-versus-temperature curves indicate that the temperature at the time of the reaction was 800 °C. Post-test examination revealed that temperatures high enough to melt the 303 stainless steel cans (1500 °C), form a Ta-Fe eutectic (1400 °C), and put the Mo into solid solution with the Fe (1400 °C) had been attained.

The exact cause of the higher temperatures is not clear at this time, but it is known that the material used for a pressure transmitting media, namely, pyrophyllite (Grade A Lava), contains 6% by weight water of hydration. It is also known that this water is released at about 600-800 °C with an accompanying increase in volume of the lava. Calculations of the heat released from a NaK-H₂O reaction at 800 °C indicate that temperatures in excess of 2000 °C could be reached depending on the rate of heat leak. Future tests will concentrate on replacing the pyrophyllite with BN and Grade 1136 Lava in an effort to isolate the possible NaK-H₂O reaction.

A high pressure form of B₂O₃ was prepared in 3.2 hrs at 39 kbars and 680 °C. The original material was amorphous B₂O₃ glass, and the high pressure material was monoclinic with $a_0 = 4.52 \text{ \AA}$, $b_0 = 4.14 \text{ \AA}$, $c_0 = 4.64 \text{ \AA}$, and $\beta = 120^\circ 25'$. This structure is 20% more dense than the hexagonal crystal structure normally stable at ambient conditions. The monoclinic phase is not as hygroscopic as the hexagonal phase although prolonged exposure to air will cause some H₃BO₃ to be formed.

The rutile structure of TiO₂ has been formed at 36 kbars and 500 °C for 0.6 hr. Normally (at 1 atm.), the rutile structure is formed only above 900 °C. It was hoped that a previously reported more dense form of TiO₂ would be formed, but only the rutile structure was evident from x-ray analysis.

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

Gas Cooled Loop Vendor Data Review. The stress report for the stainless steel heat exchanger has been reviewed by Teledyne Materials Research, and it is their opinion that the revised design submitted by Fansteel Corporation does meet the stress report requirements of ASME BPVC Section III, Nuclear Vessels, with respect to method.

The drawings for the primary heater control have been reviewed, and changes will be required to permit manual selection of either the heater exit controller or the gas loop core exit controller, instead of the automatic selection relay, as an option.

Several other contract submittals have been reviewed and the comments transmitted to the Architect-Engineer.

Gas Cooled Loop Design Review. Three packages of drawings describing the GCL pipe support arrangement were reviewed to the extent permitted by the data supplied, and comments were transmitted for distribution to Ebasco and the other approval agencies.

Other GCL drawing and specification revisions were reviewed and signed off.

The outstanding loop construction component and system operational specifications were signed at the AEC's request so they could be released for preliminary scheduling. Ebasco will incorporate PNL comments at such time as our review is complete.

Analytical Instrumentation. The inspection procedures for the analytical instrumentation being constructed by Montek, Salt Lake City, Utah, are nearly complete. It is necessary to verify that the analytical results on a known sample are within the calibration range of the instruments and that the water monitors will measure less than one ppm. This can be accomplished by the use of two or three bottles of calibration gases. The final testing will be done as follows:

1. PNL will use purified gas from the model loop purification system as a reference for the water monitors.
2. Gas mixtures will be prepared and verified with our chromatograph. The mixtures will represent a contaminated or off-standard condition with approximately 8 ppm H₂, 4 ppm O₂, 6 ppm N₂, 4 ppm CH₄, 5 ppm CO, and 5 ppm CO₂.
3. The high level CO₂ (300-500 ppm) sample will be used in conjunction with the CO₂ analyzers to test operation of the dilution train rotameter box.

The analytical instrumentation now being used on the model loop was demonstrated and explained to personnel from Idaho Nuclear. Further meetings will be planned to orientate operational personnel for the analytical instrumentation for the ATR gas loop.

Meetings. Meetings were held in San Francisco on June 27 and 28 to resolve all outstanding differences and claims in existence on the main instrument panel order.

It was determined at this meeting that welding inspection for the transmitters and ultrasonic testing on the Hoke valve bodies might be relaxed if it would improve the panel delivery. Ebasco continued to insist that their specification P-3001 was specifically applicable to the transmitter panel. All other items identified as problems by recent vendor correspondence were discussed in terms of both cost and delivery. The welding problem and results of PNL metallography were discussed, and the PNL recommendation that the vendor be requested to supply a procedure, qualification weld on tubing, and samples for metallographic examination for each weld configuration was accepted by the other agencies. It was agreed that this would be a design change, since the requirements were not explicitly called out in P-3001.

On June 26, J. Stearns, W. Thomas, and E. Heck visited LCL Controls in South San Francisco, where the main instrument panel is being fabricated. All cutouts except those for the heater controls, the M-G set switches, and the GE meters have been made, and the panel had been painted. Work on installation of the instruments and wire trays was proceeding, with wiring expected to begin the following week.

Metallic Fuels Development (G. A. Last)

Irradiation of Thorium-Uranium-Zirconium Fuel Elements

The three Th - 2.5 wt% U (93.2% U-235) - 1.0 wt% Zr tubular fuel elements being irradiated in the P-7 hot water loop in the ETR were transferred to the Test Reactor Area (TRA) hot cell for visual examination, air weighing, and dimensional measurement. The visual inspection confirmed the presence of a heavy film (presumably ZrO_2) on the outer surfaces that was cracking and spalling, mostly at the element center. Filming on the inside tubular surface appeared to be intact. The braze weld eutectic closures did not appear corroded or excessively filmed although some banding was evident within the heat-affected zone. There were no indications that localized or accelerated clad corrosion (pitting) was occurring. No indication of clad thinning or necking was seen on the surfaces of the elements. The inner tubular bores were smooth and free of any indications of mechanical instability.

The unknown extent of clad corrosion weight gains (filming) and weight losses (spalling) has created uncertainty as to the accuracy of swelling data derived from interim fuel element bulk density measurements made in the ETR basin. Air weighing of the elements revealed that corrosion weight gains and weight losses have been nearly compensating, and the current air weights are not significantly different from the initial preirradiation weights. With this being the case, the validity of interim fuel swelling data is substantiated. The following table shows a comparison between the routinely determined fuel swelling from interim weight data and fuel swelling from air cell data.

Comparison of Swelling Data
Obtained on Th-U-Zr Fuel Elements

<u>Element</u> <u>(Exposure)</u>	<u>Fuel Swelling, % ΔV</u>	
	<u>Interim</u> <u>Basin Data</u>	<u>Air Hot</u> <u>Cell Data</u>
GEH-10-64 (13,000 MWd/tonne)	1.68	2.10
GEH-10-65 (18,600 MWd/tonne)	3.35	3.29
GEH-10-71 (9,400 MWd/tonne)	1.29	1.47

Dimensional measurements indicated that the inside diameters of the tubular elements have not changed. Lengths increased quite uniformly with burnup at the rate of about 0.028 inch (for 8½-inch length) per at.% BU. Outside diameter measurements indicate that the elements are "barreling", for the diameter increases at the element center are 50 to 100% greater than they are near the end closures. Estimates of fuel volume increases made from the dimensional measurements agree closely with those determined from weighing the fuel elements in water in the ETR basin.

These three elements were returned to the P-7 loop and are continuing their irradiation. Three other elements which were being irradiated in the M-3 loop are being stored in the viewing basin at the ETR site. There is every reason to believe that the elements can successfully attain their goal exposure of 3.0 at.% BU. The element inner bores will be watched for instability and the outer clad surfaces will be routinely examined for clad thinning or pitting.

High Exposure Uranium Irradiation Test

Hollow core uranium fuel elements being tested in the M-3 hot water loop of the ETR are now in the sixth reactor cycle of irradiation. At the conclusion of this ETR cycle, number 90, the estimated accumulated exposure for the 24 fuel elements will range

from approximately 600 to 6500 MWd/tonne. The maximum fuel core temperatures range from approximately 360 °C to 540 °C. These fuel elements, 0.45-inch diameter by 6.25-inch long and clad with Zr-2, are part of an irradiation test that has been designed to operate uranium fuel rods at elevated alpha phase temperatures to burnups greater than 10,000 MWd/tonne (6×10^{20} fissions/cm³). The variables being studied in this test include fuel composition, external restraint, and internal void volume. The combined effects of the plastic character of uranium during irradiation and the restraints from the cladding and system pressure are expected to cause the uranium swelling to be accommodated by a central hole. Two uranium compositions are being used: Alloy 1, containing U + 350 ppm Fe - 800 ppm Al; and Alloy 2, containing U + 150 ppm Fe - 100 ppm Si. A fuel enrichment of 4.5% U-235 is being used to achieve the desired burnup rate (10,000 MWd/tonne in one calendar year) and temperatures. Fuel rods of both compositions were fabricated by coextrusion to 0.450-inch diameter with 0.025-inch and 0.050-inch thick Zr-2 cladding.

Additional elements have been successfully brazed and TIG welded with no indications of porosity in the braze or weld area. These elements will be shipped to the ETR as soon as final machining and autoclaving is completed.

ENGINEERING DEVELOPMENT

Neutron Flux Monitors (W. G. Spear)

Regenerative Detectors (Thermal Neutron Flux)

Continued preparations furthered progress toward the comprehensive reactor in-core test program to evaluate characteristics of the regenerative thermal neutron flux detectors. Reference radiographs and photographs were made of all ten detectors preparatory to capsule assembly. All ten detectors, including four regenerative detectors, one fission detector, one gamma detector, and four self-powered beta-current devices, are being assembled into a single test capsule for irradiation at the ETR.

Extensive laboratory tests of the electronic instrumentation system to permit accurate, comparative measurements of the operational characteristics of the detectors during the extended evaluation test will be conducted to prove out the performance of the instrumentation system. This will also supply the necessary reference data prior to installation at the ETR. Instrumentation calibrations and gamma sensitivity measurements will be taken at the PNL 3.5×10^5 Ci ⁶⁰Co gamma facility.

The regenerative detector overcomes the usual rapid burnout problem at high neutron flux levels of the U-235 detector by the use of the developed U-234 - U-235 regenerating technique. In this method U-234 transmutes to U-235 to provide replenishment of the fissioned atoms and provide long-lived proper operation during in-core use.

Microwave Detectors (Thermal Neutron Flux)

The ionization caused by charged particles from an (n,p) reaction of thermal neutrons with He-3 or by the fission fragments from U-235 or special regenerating mixture will result in a plasma. If this plasma is created within a microwave resonant cavity, the resonant frequency and cavity Q will be proportional to the plasma free electron density. Thus, the frequency shift of the cavity is related to the incident neutron flux density.

During previous experiments a significant reduction of sensor sensitivity was noted. As reported, the inclusion of a dielectric capsule within the microwave cavity will reduce the sensitivity by a factor of about 10 as compared with a dielectric-free sensor cavity. Thus, it is important to obtain a sensor which is gas-tight and also allow efficiency coupling of microwave energy. Techniques to obtain suitable metal-to-ceramic seals in waveguide sections have been successful. Techniques to successfully attach these to a cavity have been partially successful to date. One assembly has been assembled at relatively low temperature (185 °C), making complete "bake-out" difficult. Other assemblies fabricated offsite contained gas leaks in the metal-ceramic seals. This failure is traceable to an incorrect assembly and brazing sequence. The manufacturer used two brazing operations, the second of which was to attach an end cap to the already assembled vacuum tight waveguide cavity body assembly. The resulting strain resulted in eventual fracture of the ceramic.

A slight change in design to overcome some of the encountered difficulties in the strain on the ceramic should allow the fabrication of satisfactory cavity sensors for subsequent in-reactor neutron flux measurements.

Regenerating Detectors (Fast Neutron Flux)

To achieve rugged, long-lived reactor in-core fast neutron detectors, it appears that suitable regenerative techniques may provide appropriate performance characteristics to achieve fast neutron flux measurements under anticipated reactor operating conditions such as severe temperatures, high fast flux densities, and high gamma levels.

The capture and fission characteristics of possible candidate isotopes are under investigation. Computer analysis methods are being used to explore the pertinent cross sections of over 20 fertile or fission isotopes. From this and possible additional data, the proper combinations of isotopes to develop a fast neutron regenerative detector will be established.

Beta Current Generator Detectors (Fast Neutron Flux)

The (n, α) reactor of Be-9 (0.1 barn cross section at 3 MeV) shows promise as possible detector material for measurement of fast flux under difficult environmental conditions. The resulting beta particle emission (3.5 MeV) occurs with a relatively short half life to 0.8 second. The beta emission generated a signal proportional to the neutron flux density. A signal current of approximately 1×10^{-6} A from one cm^3 of Be-9 is expected when exposed to fast flux of 10^{15} nv. The Be-9 in a metal oxide state can be encapsulated in a suitable sheath which is opaque to alpha particles but allows the energetic beta particles to pass through. This loss of charge from the center results in signal current. It is anticipated that balanced, twin lead, twin chamber concept as developed during thermal flux detector investigations will prove useful in avoiding interfering signals induced by gamma reactions with the cables.

Evolved Gas Detection Concept (Fast Neutron Flux)

In this unique concept for advanced reactor fast flux detection, an evolved gas released in (n, α) or (n, p) reactions with B-10 or other materials would be collected through a simple evacuation tube connected to the detector assembly. Gamma interference could be minimized by locating all instrumentation external to the reactor core. Calculations indicate that about 10^{11} atoms sec^{-1} of helium gas could evolve from one gram of B-10 in an incident fast neutron flux of 10^{13} nv. Many methods could be used to measure the evolved gas including mass spectrographic or optical spectrographic methods or even possibly a microwave resonant cavity.

Microwave Detectors (Fast Neutron Flux)

By extending the microwave techniques being developed for use in thermal neutron detection, it is anticipated that reactor fast flux may be measured. The cavity would contain a material with an appreciable cross section to fast flux such as U-238, or the proton recoil in hydrogen could be used as a source of ionizing particles. The use of hydrogen would allow replenishment of the gas as necessary. Gamma-caused ionization will prove troublesome and compensation techniques will be necessary, such as using two cavities where one is sensitive only to gamma and measure the difference in signal frequency between the two cavities.

Other techniques being considered include the use of a gas emitting material such as Be-9, wherein the gas is evolved into or collected by a microwave cavity. The resonant frequency is a function of the number of gas molecules within it, thus the frequency shift becomes a measure of the neutron flux density. It has been shown theoretically that measurements of fast neutron flux down to 10^{11} nv could be achieved for a gas evolution rate of 10^{-2} atoms sec^{-1} nv^{-1} .

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

Microwave Detection of Impurities in Coolant Gases

An atmosphere, corrosive to reactor components, is created when a reactor coolant gas is contaminated with water vapor. To detect excess amounts of water vapor in gas coolants, several microwave detection schemes are being investigated. The propagation properties of electromagnetic waves in microwave structures are affected by the dielectric constant of the propagation medium. Because the dielectric constant of a gas mixture is a function of the number of molecules of each gas present and their molecular polarizabilities, the addition of water vapor to an otherwise pure gas changes its dielectric constant.

By detecting a change in the resonant frequency of a cavity through which the gas mixture flows, an effective impurity sensor is devised. During first experiments, a detection sensitivity of 125 ppm of water vapor in helium at 300 °C and of 950 ppm at 436 °C was achieved. Time response of the detector is short, being limited by the time constant of the electronic measuring equipment. Greater sensitivity can be achieved by using a phase locked microwave oscillator and heterodyne techniques. Also being considered are methods of selectively separating the gas mixture, thereby increasing the probability of detecting a water vapor content less than the threshold detection level for a uniformly distributed gas mixture.

High Temperature Measurements

Because of its relation to reactor power level, it is required that temperature of in-core components be measured accurately and precisely. Economically, it is beneficial if temperature sensors and associated components are long lived. Several methods appropriate to the measurement of reactor temperatures using microwave techniques are being investigated.

One method, which depends upon the shift-of-frequency of a microwave cavity as temperature changes, yielded a nearly linear temperature-frequency response of 300 kHz/°C during initial experimental tests to 1000 °C. However, higher temperature capability is desired for monitoring component temperatures within new gas cooled reactors. Fabrication of several waveguide-cavity assemblies of a high melting point (1450 °C) chromium-nickel steel should allow additional experiments to be performed to about 1400 °C.

Another microwave method of measuring temperature employs a radiometer which measures the electromagnetic energy emitted from any heated body. During preliminary tests using a load with an average emissivity of about 0.8 in the frequency band of interest, a nearly linear output signal-temperature response was obtained for

temperatures to 700 °C. Sensitivity of the apparatus was 0.815 mV/°C. Using a prepared load which more nearly approximated a black body, temperature to 950 °C was measured with an increased sensitivity of 0.175 mV/°C.

Because of the linear relation between radiometer output signal and temperature, it is assumed that emissivity of the sensor black body remained nearly constant. Additional experimental work will concern the direct measurement of temperatures of materials immersed within the furnace. These tests will employ a microwave receiving antenna which intercepts the emitted radiation from the monitored object.

Infrared Techniques

In preparation for making calibration tests of the second experimental hygrometer, an excessive 60 cps noise was noted, but changes reduced this to an acceptable level. Changes previously made in the servo-driver wedge-filter design necessitated corresponding changes in the filter position indicator. A new electronic circuit and several mechanical modifications served to solve the problems.

The hygrometer has a phase-changing circuit which switches the field windings of the servo motor to proper polarity regardless of the lock-in phase of the synchronous motor driving the filter wheel. An added latching relay served to provide fully automatic operation.

Further consideration of methods of incorporation of a CO₂ absorption path into the system resulted in the simple solution of filling the sealed chamber containing the motors, lamp, and filters with dry CO₂. This procedure eliminated need for an external cell to preclude sensitivity to CO₂ in the gas being monitored.

PLUTONIUM UTILIZATION PROGRAM (F. G. Dawson)

Fuels Development

PRTR Testing

Recent fuel rod examinations associated with the PRTR power calibration have revealed fuel melting at an indicated maximum rod power generation of 20.0 kW/ft. One of the two rods melted to approximately 10% of the radius; the other rod melted to approximately 33% of the radius. Both rods were presumably irradiated under the same conditions. The reason for the difference in the indicated melt radius is not understood. Based upon an extrapolated actual power to produce melting of 20.5 kW/ft, fuel temperatures at

20.0 kW/ft should have been slightly below melting. The most recent postirradiation examination results and an analysis of the observations based upon these results are summarized as follows:

Rod No.	Indic. Max. Rod Power (kW/ft)	Est. Act. Max. Rod Power (kW/ft)	Approx. Time at Max. Power (hours)	Col. Grain Growth Radius (%r)	Indic. Temp.* Grain Assumed 1800 °C	Max. Fuel Growth is When Col. Occur @ 2000 °C	Calc. Max. Fuel Temp. Assuming 20.5 kW/ft to Melt (°C)
FS-03	16.8	17.8	48	50	2140	2360	2590
FS-19	17.0	18.0	>86	60	2330	2560	2600
FR-31	17.9	19.0	80	60	2330	2560	2700
FR-45	18.6	19.7	40	64	2460	2640	2750
FR-47	20.0	21.2	>68	70	2600	>Melting	2875
FR-29	20.0	21.2	>68	76	>Melting	>Melting	2875

*Derived from calculated fuel temperature profiles which indicate the onset of fuel melting at 20.5 kW/ft.

From these observations it appears that the indicated power to produce fuel melting is between 19.4 and 20.0 kW/ft and probably closer to 19.4 than 20.0 kW/ft. The accuracy of reactor instrumentation is being evaluated to experimentally establish the actual power (estimated to be approximately 20.5 kW/ft) to produce the onset of fuel melting.

Destructive examination of the end regions of the two rods that underwent fuel melting revealed that the depleted UO_2 pellets effectively prevent excessive fuel temperatures in the end-cap regions. There is no evidence of pellet-fuel interaction and only minor pellet cracking has occurred. Excessive fuel temperature in the end cap regions was an operating limitation in the past. Efforts are being initiated to better define the conditions within the core so that less operating conservatism is required and actual fuel operating conditions are known.

Long-term fretting and wear tests are continuing with a prototype 8-rod FERTF test element under PRTR coolant conditions in the out-of-reactor EDEL test loop. The element has been exposed 63 days without evidence of wear between components or between the element and the pressure tube. The behavior of a loose circumferential strip band around the rod bundle has been satisfactory.

A summary of a paper, titled "Plutonium Fuel Irradiations in PRTR - Present Status," by M. D. Freshley, T. B. Burley, and S. Goldsmith was submitted for possible presentation at the forthcoming Winter Meeting of the American Nuclear Society.

PRTR Pellet Rod Procurement

One commercially fabricated PRTR fuel rod containing cold pressed and sintered mixed oxide pellets was received from the vendor. Nondestructive examination of the rod was started.

Two additional rods of identical design are expected to be delivered in early August 1967. Extra pellets will be delivered with the remaining two rods so that the fuel material can be fully characterized.

All three rods will be used to compare commercially designed and fabricated pellet rods with BNW-designed and fabricated powder rods. Hot pressed pellets as well as cold pressed and sintered pellets will be used in the comparison.

Instrumented Fuel Rods

Four fuel rods instrumented to measure fuel rod gas pressure and temperature continue to operate satisfactorily in PRTR.

Two of the fuel rods have operated at maximum linear power ratings of approximately 20 kW/ft and two have operated at maximum linear power ratings of approximately 10 kW/ft. Calculated exposures achieved by the rods are 1280 MWd/tonne and 594 MWd/tonne, respectively. The gas-pressure buildup in the high power fuel rods is approximately three times that in the low power fuel rods (56 psi versus 17 psi, respectively). In both sets of fuel rods, gas release (fuel rod gas pressures) increases in a stepwise manner during reactor startups, then levels off or decreases slightly during steady state operation.

ETR Testing

An intentionally defected PRTR prototype 7-rod cluster element containing vibrationally compacted enriched UO_2 fuel was successfully irradiated in the ETR P-7 pressurized water loop at a maximum linear rod power generation of 28 to 29 kW/ft. This power generation resulted in an estimated 60 to 65% of the radius molten at the plane of the defect. The defect consisted of a 1/16-inch-diameter hole drilled through the Zircaloy cladding. After irradiation under these conditions for nine effective full-power days, the element was discharged from the reactor for postirradiation examination.

During its residence time in the reactor the defected element was subjected to three programmed power increases, two of which were from zero power. The element was also subjected to one scram recovery which included a very rapid power increase (3 min) from 1 to 50% of full power. Although the activity release to the loop piping was quite high during the scram recovery, modest amounts of activity were released during the programmed power increases. Increases in

reactor power and coolant temperature caused activity bursts which decreased to low equilibrium values during steady state operation.

A cursory examination of the defected rod in the ETR basin did not reveal any physical changes as a result of irradiation. A detailed postirradiation examination will be conducted to compare the performance of the defected rod with the performance of adjacent nondefected rods. The defected rod will also be examined for evidence of dimensional changes, waterlogging, fuel washout, fuel relocation, fission product and/or plutonium migration, and stoichiometry changes.

TREAT Transient Testing

Discussions with ANL have indicated that more than five months will be required for design, construction, and testing of an instrumented piston autoclave for use in transient testing of fuel rod clusters in TREAT. To avoid a delay in the program, a simple, low cost autoclave design capable of accommodating both single and multiple fuel rods and which incorporates suggestions made by TREAT Facility personnel, is presently being prepared. It is anticipated that single rod transient experiments, utilizing the simple autoclave, will be conducted by September 1967.

Additional discussions with TREAT Facility personnel indicate that transient testing of multirod clusters in a recirculating coolant are feasible. Various designs of an autoclave with recirculating coolant capabilities are being investigated.

Two high-speed motion pictures of vibrationally compacted UO_2 (5.01 at.% U-235) fuel rods undergoing transient irradiations in the TREAT Facility were obtained. One rod was subjected to a total transient energy input of 335 cal/g UO_2 , and the other rod to a transient energy input of 418 cal/g UO_2 . The cladding near the center of both fuel rods appeared to fail first, followed by fragmentation of the rods into small particles and fines. Neither rod displayed the well defined high temperature bands that occurred with a vibrationally compacted UO_2 (9.54 at.% U-235) fuel rod previously subjected to an energy input of 357 cal/f UO_2 .

Examination of x-ray films of one vibrationally compacted UO_2 (9.54 at.% U-235) fuel rod that was subjected to threshold failure of 241 cal/f UO_2 suggests that initial clad penetration is aided by the release of entrained gases in the fuel at elevated temperatures. The x-ray film shows a number of propagating separations in the fuel column near the center of the fuel column. Offgas analyses of fuel obtained from history samples indicates that the entrained gas release as a function of temperature is approximately 1.3×10^{-4} cc/g fuel/ $^{\circ}\text{C}$ in the range of 900 to 1650 $^{\circ}\text{C}$.

Project CAB-002

Installation of the vacuum welding chamber has been completed except for control wiring. Physical completion of the project should be in early August.

Underwater Profilometer

All equipment for the underwater fuel element handling facility has been received. The project will be closed out in August.

High Exposure Plutonium Studies

A 63-gram sample of PuO₂ was prepared from Dresden-product plutonium nitrate obtained from Nuclear Fuel Services. The PuO₂ had a surface dose rate of 2.73 rad/hr as read through 0.033 inch of polyethylene. This dose rate is slightly lower than Shippingport oxide rates as expected, in view of the somewhat lower Pu-241 contents (7.6% versus 6.9% Pu-241 for Dresden).

Gamma and x-ray attenuation was measured through various materials from recycled plutonium containing about 10% Pu-240. One-half inch of lucite lowered the gamma dose by a factor of three, and 30-mil lead-loaded gloves reduced the gamma dose by a factor of five. The thicknesses required to reduce the gamma dose by factors of two and ten are given below (with an extrapolated correction for the three layers of single plastic sheet covering the PuO₂ powder source).

<u>Material</u>	<u>Thickness Required to Reduce Dose by Factor of 2</u>	<u>Thickness Required to Reduce Dose by Factor of 10</u>
Lead	-	12 mils
Stainless Steel	-	55 mils
Lead-Loading Gloves	-	50 mils
Material		
Neoprene Glove	10 mils	-
Material		
Lucite	200 mils	-
Vinyl Plastic	500 mils	-

Neutron attenuation was also studied. Using a plutonium fluoride neutron source the thickness required for a twofold flux attenuation was: 3 inches (a combination of 2½ inches) Lucite and ½ inch Boron-polyethylene; 2½ inches for Boron-polyethylene and 2½ inches for Paraffin. Six and one-half inches of paraffin are required for a tenfold flux attenuation. The attenuation properties of paraffin and low density polyethylene should be similar for neutrons.

The neutron dose rate attenuation will be significantly different from the neutron flux attenuation. How much different is not known since the neutron energy spectrum is unknown after the neutrons pass through the attenuating materials. The dose rate is, however, very energy dependent. The dose per unit fluence changes by a factor of about 17 between 1 keV and 10 keV, and the dose per unit fluence changes by a factor of about 25 between 10 keV and 1 MeV.

Isotopic analyses have been completed on the Yankee fuel samples. One Dresden composite fuel sample from Nuclear Fuel Services plutonium product is being analyzed.

Reactor Physics

D₂O Moderated Systems

Batch Core Experiment in the PRTR. The performance and analysis of batch core power tests in the Plutonium Recycle Test Reactor (PRTR) have continued. Power Test No. 6, Transient Fission Product Poisoning, has been completed, and the results are being analyzed. The boron concentrations in samples of PRTR moderator which were collected during the test have been determined using the Thermal Test Reactor (TTR). The first two samples, which were expected to be identical, showed a difference of 0.16 wppm B-10. The concentrations of the remaining samples were about as expected. The measured reactivity transient was not as large as had been calculated using the XRST code. A study of the possible causes of the discrepancy is in progress.

A summary, entitled "Power Tests with the UO₂ - 2 wt% PuO₂ Batch Coore in the PRTR," by J. W. Kutcher, R. E. Harris, and R. I. Smith, has been submitted to the American Nuclear Society for presentation of a paper at the 1967 Winter Meeting.

Technical Assistance to PTU. Reactor physics assistance has been furnished to the Processing Technology Unit of the PRTR Section, in the establishment of operational limits for the batch core in the PRTR which will assure that no fuel melting occurs. These efforts include modification to PRTR Test 136 (Determination of Allowable Fuel Element Rod Power), reevaluation of measured and calculated axial flux peaking factors, and the determination of the need for and the planning of additional tests as necessary.

Effect of Cs-137 Half-Life and Fission Yield Uncertainties on Cross Section Ratios Derived from Burnup Data. The contribution of the cesium-137 half-life and fission yield uncertainties to the uncertainties of the cross section ratios obtained from a least squares fit of PRTR plutonium-aluminum burnup data has been determined. This contribution must be calculated independently from the LIKELY least squares uncertainty calculation. Just like any other least

squares fitting analysis, LIKELY assumes that the data points are independent measurements. Thus, the uncertainties of the Cs-137 half-life $T_{1/2}$ and fission yield Y which systematically affect all data points must be excluded from the LIKELY analysis. Cs-137 enters the analysis because it was used in establishing the pre-irradiation plutonium content of the samples. The half-life and fission yield occur in the combination $T_{1/2}/Y$. Since the experimental data (the plutonium isotopic concentrations) are systematically affected by the value of $T_{1/2}/Y$, the cross section ratios X derived from this data are also a function of $T_{1/2}/Y$. Thus

$$\begin{aligned}\sigma_X^2 &= \left(\frac{\partial X}{\partial T_{1/2}} \sigma_{T_{1/2}} \right)^2 + \left(\frac{\partial X}{\partial Y} \sigma_Y \right)^2 + \sigma_L^2 \\ &= - \left(\frac{Y}{T_{1/2}} \frac{\partial X}{\partial Y} \sigma_{T_{1/2}} \right)^2 + \left(\frac{\partial X}{\partial Y} \sigma_Y \right)^2 + \sigma_L^2\end{aligned}\quad (1)$$

where σ_L is the one standard deviation uncertainty in X as determined by the LIKELY fit to the data. The quantity

$$\frac{\partial X}{\partial Y} = \frac{\Delta X}{\Delta Y} = \frac{X_2 - X_1}{Y_2 - Y_1}$$

was determined by making LIKELY least squares fits to two sets of data. Set #1 was computed using $Y_1 = 0.0648$ and set #2 used $Y_2 = 0.0672$.

Using $T_{1/2} = 29.68 \pm 0.30$ yr and $Y = 0.0648 \pm 0.0019$, the results of Equation 1 are shown in the table.

Effect of Cs-137 Half-Life and Fission Yield
Uncertainties upon the Cross Section Ratio Uncertainties

	<u>X</u>	<u>$\Delta X/\Delta Y$</u>	<u>σ_L</u>	<u>σ_X</u>
α^{239}	0.4391	7.50	0.0032	0.0154
$\frac{\sigma_a^{240}}{\sigma_a^{239}}$	0.5789	6.00	0.0058	0.0134
α^{241}	0.3513	4.21	0.0127	0.0153

The cross section ratio $\sigma_a^{240}/\sigma_a^{239}$, which varies with exposure, is evaluated at zero burnup. α^{239} and α^{241} are the Pu-239 and Pu-241 capture to fission ratios. The table makes it clear that the

uncertainties in $T_{1/2}$ and Y contribute heavily to the uncertainties in the cross section ratios. Thus, improved cross section ratio values await better values of $T_{1/2}$ and Y .

Thorium Loading in the PRTR. A burnup calculation was performed using the ZODIAC code for a PRTR loading of 19 UO_2 - 2 wt% PuO_2 ceramic fuel elements surrounded by 66 Th - 3.7 wt% U-235 metallic fuel elements. The ceramic elements were assumed to have an average exposure of 13,000 MWd/tonne, and the metallic elements were unirradiated. The lifetime of this core was calculated to be 300 days at a 70 MW power level.

A summary of the physics calculations and results for a thorium loading in the PRTR has been written and will be included as an appendix of a document covering all aspects of a thorium loading in the PRTR.

H₂O Moderated Systems

Reactor Application Studies. The design of the core hardware to be used in a joint USAEC-CNEN cooperative program to study the problems of introducing plutonium in H₂O reactors is essentially complete. Fabrication of the core components is now in progress. A total of 600 2.3% enriched UO_2 fuel elements are being modified for use in this core. The elements will be reduced from an active length of 60 inches to 36 inches. Control rods with fuel followers are being adapted to this core design. The rods are designed to accept either the UO_2 or the mixed oxide fuel as followers. Completion of core hardware and fuel modification is expected in late August.

UO_2 - 4 wt% PuO_2 Fuel Rods. Fabrication of the UO_2 - 4 wt% PuO_2 fuel rods was completed on schedule by the end of June. Preparations are nearly finished to start approach to critical experiments in the Critical Approach Facility (CAF). Initial measurements will be in the 0.8 inch hexagonal lattice. This work has been delayed by a lack of craft labor to complete the CAF modification and improvement program.

Gamma Scanning of EBWR Fuel Pins. The scanning of 17 EBWR fuel pins with the PNL gamma scanner to determine the relative distribution of fission products has continued during the past month. The work has been hampered by changes in ambient temperature which caused difficulties with the analyzer electronics. These problems have now been corrected.

Six different fuel compositions are available for scanning, and they are (1) Natural UO_2 , (2) PuAl 8% Pu-240, (3) PuAl 26% Pu-240, (4) PuO_2 - UO_2 26% Pu-240, (5) PuO_2 - UO_2 20% Pu-240, and (6) PuO_2 - UO_2 8% Pu-240. The PuAls were initially 3.35 wt% Pu in aluminum and the PuO_2 - UO_2 's were 1.50 wt% PuO_2 in depleted UO_2 . Fuel composition (6) is the normal plutonium enriched fuel for the EBWR burnup experiment and surrounded the other five special compositions in the core.

The results to date have shown that the power is peaked at 11 inches above the bottom of the fuel, and there is little burnup in the top 10 inches of the fuel.

Decay Scheme Analysis. A Nuclear Data Model 2200 multichannel analyzer and 20 cm³ Ge(Li) coaxial detector and a TMC 4096 multi-parameter analyzer and 15 cm³ Ge(Li) coaxial detector were used to study the decay schemes of Lu-176m and Zr-97 - Nb-97 - Mo-97.

A high-energy gamma-ray analysis of Lu-176m has confirmed the existence of two low-intensity gamma rays near 1060 and 1160 KeV reported earlier. In addition, three previously unreported weaker intensity gamma rays were found in the same energy region. All five gamma-ray photopeaks were found to decay with a half-life near that of Lu-176m. An intensity and energy analysis is currently under way.

A gamma-ray analysis of the Zr-97 - Nb-97 - Mo-97 mixture has confirmed the previously reported close-lying double-photopeaks in the 1024 and 1280 keV energy regions. A second analysis was conducted on chemically separated zirconium and niobium. The separation scheme, modified from a procedure by Jurriaanse and Morre (Anal. Chem., 38, 964 (1964)), resulted in a 99.95% pure niobium source. As a result it was shown that in each of the above cases, one of the gamma rays belongs to Nb-97 and the other to either Zr-97 or Nb-97m. A further analysis of available data may result in the definite assignment of these gamma rays to the decay scheme. In addition, five previously unreported low-intensity high-energy gamma rays were found that seem to support some of the energy levels tentatively assigned to Mo-97 earlier.

Calculation of Fission Product Production in Irradiated Fuel. A set of differential equations has been derived which describe the production of selected groups of fission products in fuels enriched with uranium and/or plutonium while under irradiation in a thermal reactor. The equations have been solved for the atom densities of the selected fission products as functions of irradiation conditions and cooling times. Attempts will be made to correlate the results from these calculations with the results from high resolution gamma spectroscopy of irradiated fuels.

Temperature Coefficients of PuO₂-UO₂-H₂O Lattices. A summary of a paper, entitled "Temperature Coefficients of PuO₂-UO₂-H₂O Lattices," has been submitted to the ANS for presentation of the paper at the Winter Meeting of the ANS. The paper will give analytical correlations for temperature coefficients measured in a number of PuO₂-UO₂-H₂O lattices.

Irradiated Fuel Shipment. The second shipment of irradiated fuel was made from ANL to PNL. These fuel pins (17 total) were removed from the plutonium zone of the EBWR around the first of May. The average exposure of these pins is about 2300 MWd/tonne. The

transfer of this fuel from the Yankee cask to the PRTR cask was made in the 100-D Area basin. The fuel is presently in the PRTR basin where gamma scans of each pin will be performed.

The EBWR power operation was terminated on June 29. Post-irradiation experiments to determine boric acid worths, control rod worths, and k_{eff} for the plutonium zone are being conducted. The final shipment of irradiated fuel from ANL to PNL will be made sometime in August or September.

Commercial Fabrication of PRCF Fuel Rods

Critical experiments in the PRCF require a large quantity of (4 wt%) PuO_2 - UO_2 test fuel rods. Commercial procurement is nearing completion under the two-step procurement procedure to obtain the fuel rods.

Seven possible commercial fabricators were informed of the proposed work. Five of the possible seven fabricators responded with technical proposals under the step-one procedure. Areas of technical variance in these proposals are being resolved with these companies. Request for pricing of fuel rods is planned with notification to fabricators under step-two planned for release about August 4. About 200 test rods are being requested for delivery on November 1, 1967; other options are included about possible delivery dates for additional fuel rods.

During discussion of variance in technical proposals with commercial fabricators several items were modified or additionally defined on the original PNL proposed specification to help meet present industrial capabilities. For example, the original fuel design provided for either packed-particle or pelleted fuel. Since all companies responded with the pelleted-fuel option, a new fuel rod design drawing was completed with simplified details to provide for pelleted fuel. Smear-density tolerances were also increased to $(86.0 \pm 2)\%$ from $(86.0 \pm 1)\%$.

The core is now defined as 4.00 wt% PuO_2 (4.04 MAX - 3.96 MIN) in UO_2 (natural isotopic) as PuO_2 - UO_2 pellets to give a fuel rod smear density of $(86.0 \pm 2)\%$ theoretical density per lineal inch of fuel. Density variations of pellets are to be $\pm 2\%$ maximum. The test of plutonium homogeneity was also defined as a combination of qualitative alpha autoradiography and quantitative wet-chemical analysis of pellets randomly selected as defined by an approved sampling plan. Additional changes involved use of a UO_2 pellet spacer between fuel and bottom end cap, welding conditions, and additional use of ASTM specifications for Zircaloy-2 tubes and end caps.

PuO₂ Particle Size Studies

Two types of half-inch diameter PuO₂-UO₂ rods were investigated at various square lattice pitches as possible candidates for the initial PCTR water lattice experiments; low exposure (LX) of 0.90 wt% PuO₂ content with 7.24 at.% Pu-240 plutonium and 0.234 at.% U-235 uranium, and high exposure (HX) of 2.69 wt% PuO₂ content with 29.34 at.% Pu-240 plutonium and 0.182 at.% U-235 uranium. Six lattice spacings from 0.65 to 1.15 inches (moderator to fuel volume ratios of 0.87 to 5.46) were studied. Infinite lattice, cell averaged, thermal group constants were obtained from modified THERMOS using an isotropic outer boundary condition and the standard approximate correction for anisotropic scattering in water. Fast group constants were obtained from HRG using resonance parameter input from the PANCOF code.⁽¹⁾ The Dancoff-Ginsburg method without a clad correction option was used in this code with the shadowing by the nearest ten rings of neighboring rods taken into account. A short summary of the primary results of these infinite lattice mixed oxide calculations are contained in Table V.

Preliminary 4-group, 1 dimensional diffusion theory calculations using HFN were made to predict both the critical number of driver columns for the PCTR with all control rods open and the degree of spectral equilibrium within the test lattice when the 1.00 inch square pitch low exposure mixed oxide is loaded in the water tank assembly. The critical number of drivers was calculated to be 21.7 with both the inner and outer tank loaded (216 rods) and 32.7 with just the central tank loaded (104 rods). These predicted critical driver column numbers have been corrected to account for errors between the calculated and measured driver column and open control rod worths in this "first shot" PCTR model. This model is presently being refined to increase the accuracy of these calculated worths for future test lattice calculations. The degree of spectral equilibrium in the test lattice is determined by the behavior of the fast-to-thermal flux ratio, which should remain reasonably constant over the central few lattice units and should compare with that calculated for the infinite lattice. In the case where the central tank only was loaded, the fast-to-thermal flux ratio was calculated to be 1.6184 at the center of the tank. This compares quite favorably with the value of 1.5895 determined for the infinite lattice. This ratio is also quite constant, decreasing only 1% at a radius of 4.9 cm from the center of the tank, which includes about 12 rods plus their associated moderator (the radius of the central tank is 14.7 cm). The fast-to-thermal flux ratio was constant over a larger area and slightly higher, 1.6245, with both tanks loaded.

(1) G. L. Simmons and W. L. Purcell, "Comparison of Methods for Obtaining Dancoff Correction," BNWL-CC-1099, March 1967.

Calculated Infinite Lattice Parameters

<u>Low Exposure Mixed Oxide (0.90 wt% PuO₂)</u>					<u>High Exposure Mixed Oxide (2.69 wt% PuO₂)</u>			
<u>Lattice Spacing (in)</u>	<u>$\overline{\eta f}$</u>	<u>f</u>	<u>ϕ_{fast}/ϕ_{th}</u>	<u>k_{∞}</u>	<u>$\overline{\eta f}$</u>	<u>f</u>	<u>ϕ_{fast}/ϕ_{th}</u>	<u>k_{∞}</u>
0.65	1.5126	0.9356	6.8135	1.0886	1.5847	0.9658	14.1197	1.0740
0.80	1.3839	0.8554	2.8910	1.1716	1.5200	0.9159	5.2846	1.2201
0.95	1.2458	0.7700	1.7912	1.1255	1.4261	0.8556	2.9876	1.2379
1.00	1.1999	0.7416	1.5895	1.0988	1.3904	0.8341	2.5828	1.2287'
1.05	1.1456	0.7136	1.4303	1.0692	1.3558	0.8120	2.2680	1.2145
1.15	1.0667	0.6593	1.1968	1.0049	1.2828	0.7676	1.8152	1.1752

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From these results and other considerations, the 1.00 inch pitch LX lattice was chosen for further study.

The fast group parameters for this lattice only were recalculated using the Sauer rather than the Dancoff-Ginsburg method of obtaining the Dancoff factor. However, the effect of this change was almost negligible. k_{∞} was recalculated to be 1.0993, an increase of only 0.5 mk and the infinite lattice fast-to-thermal flux ratio became 1.5889 as compared to the previously calculated value of 1.5895.

BNWL-522

Code Development*

Scattering Kernel for Water. Codes for generating scattering kernels for water from a spectral function composed of a sum of gaussians currently appear to operate properly. Since such a technique requires numerically performing a double integration, two separate approaches depending on which integration is performed first were investigated. Both approaches are feasible and produce results that agree within the expected numerical accuracy of the codes. Experimental data are being collected to fit the parameters required in the spectral function. When this is complete, scattering kernels will be generated for reactor analysis use.

BNW Master Cross Section Library. All of the isotopes that have been modified or added to the BNW Master Library since it was published in 1965 (BNWL-CC-325) have been printed on duplimat paper and will be published shortly as a supplement.

ENDF/B Neutron Cross Section Library. Several important isotopes used in reactor physics calculations were listed as containing serious but easily correctable errors. These errors have been corrected, and the production of cross section libraries for THERMOS, HAMMER, and MC² has been attempted. Several of the processing codes contain errors that prevent the use of the ENDF/B Library until the processing codes can be successfully debugged.

Programs GAND, GAF, and GAR. The General Atomics computer codes GAND, GAF, and GAR have been received and are being converted for use at BNW.

GAND produces GAF and GAR cross section libraries from the ENDF/B Neutron Cross Section Library. GAF is the latest version of GAM-III and is used to calculate neutron spectra and averaged cross sections in the high energy region. GAR is used in the evaluation of leakage-dependent resonance absorption.

Program EDIT. EDIT, a program which reads the ENDF/B neutron cross section library tape, was updated to treat one of the options in the fifth file. This file contains the energy distribution for exiting particles. The output formats were changed to increase the number of significant figures that could be punched. This was necessary so that the punched card output will duplicate the library data in all cases.

Program ETOM-1. ETOM-1, a program that calculates MUFT4 or MUFT libraries from the ENDF/B library, was updated from a list of corrections received from the Brookhaven evaluation center. A run requesting the isotopes niobium and tantalum was made. It was found

*Partially supported by Douglas United Nuclear and General Electric N-Reactor Department.

that the program cannot properly handle a resonance with energy less than 6.2481 -Olev. Since bound state resonances ($E < 0$) are less than this limit, Tantalum-181 cannot be processed. ^CAdditional difficulties were encountered that prevented Niobium-93 from being processed. It appears that any isotopes having resonance parameters cannot be processed by ETOM until additional debugging has been completed.

Program DAFFY. A computer program entitled DAFFY is being written that allows increased precision in the determination of gamma-ray energies, their associated errors and intensities from pulse height analysis data. It is unique in that it considers the errors for the gamma-ray photopeaks of the unknowns and the calibration sources, the calibration energy errors, and the analyzer linearity are considered. Program LIKELY, capable of handling least squares analyses when errors are associated with both the X and Y parameters, forms the basis for the program and is used to determine the calibration equation. Sub-routines are added that determine the average channel for each gaussian-shaped photopeak, the average background or compton level under each photopeak, and the net intensity under the photopeak. Other than control cards, the input data is used directly as it is compiled on a multi-channel analyzer. The output of the program includes usual gamma-ray analysis information as well as gross and net plots of the photopeaks and their respective backgrounds. The program is flexible in that plotting and intensity calculations may be accomplished without the energy determination or visa-versa.

Space-Time Kinetics. Efforts are under way to have available techniques at BNW representative of the present state of the art in the area of space-time neutron kinetics. A first goal is to have the WIGLE and WIGL2 kinetic codes available on site. A second goal is to develop competence in the more standard analytical methods used in kinetics studies; i.e., the point kinetic, adiabatic, modal, and nodal methods. The WIGLE codes have been ordered, and it is anticipated that there will be no great difficulties in converting the codes over to the Univac 1108 machine. Work is also underway to develop the codes and techniques necessary to accomplish the second goal mentioned above.

RBU Monte Carlo Code. A rough draft of the document entitled "Results of Cell Calculations using the RBU Monte Carlo Code," has been written. Also, the writing of a rough draft of a document entitled "Fortran Service Codes for the RBU Monte Carlo Code" is approximately one-third complete.

A literature search is being conducted to determine the available Monte Carlo reactor physics codes. The purpose is to determine the direction to be taken in the development, what further work needs to be done to develop Monte Carlo techniques at PNL and also what

new techniques or developments to incorporate in the RBU code.

Program DRAFT. A computer program, DRAFT, is being developed which is designed to help analyze fission product data from uranium and/or plutonium fuel elements with the expectation of determining absolute burn-up rates with nondestructive techniques.

The program considers the rate of production by U^{235} , U^{238} , and Pu^{239} . In addition, the rate of decay and depletion, where applicable, are taken into account.

Any two isotopes are studied simultaneously. Plots of their concentrations, as a function of yield and flux, are obtained upon option.

Equations for the isotopes in question have been derived and programmed for use on the UNIVAC-1108.

Cycle Analysis

ALTHAEA Libraries

A new standard library (14 isotopes) and a new americium-curium library (19 isotopes), is now available. These libraries incorporate the latest available updated isotope data on the BNW master library.⁽¹⁾ The updating here involves not only the addition of new and more detailed cross section data but a more accurate way of describing this data.⁽²⁾ Both libraries represent a normal temperature range from zero to 600°C. The americium-curium library differs from the standard library only by the additional americium and curium isotopes. This makes the standard library favorable for rapid reactor surveys, whereas the americium-curium library will allow a study of the production of higher isotopes.

The U-233, U-235, and Pu-239 data was taken from the work of Westcott et al.⁽³⁾ The Pu-241 data is an old version recommended by Phillips Petroleum favoring a lower capture cross section which provides a lower α -value.

A very high temperature library is now available. This covers a

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- (1) K. B. Stewart, "BNW Master Library," USAEC Report. BNW-CC-325, September 1965. BNWL-CC-325 Rev. June 30, 1967, to be published.
- (2) Monthly Report of Advanced Concepts and Analysis Section of Mathematics Department, April 1967.
- (3) C. H. Westcott, et al. "A Survey of Values of the 2200m/sec Constants for Four Fissile Nuclides," Atomic Energy Review IAEA, Vienna III 2(1965).

range from 1500°C to 3500°C. The same updated data was incorporated in this library as that used for the above mentioned libraries. The upper cut off of the epithermal group is at 3 eV, whereas the cut off for the standard and americium-curium libraries is taken at 0.68256 eV. It was necessary to divide the temperature range into two groups (1500°C to 3000°C to 3500°C) because of a problem in obtaining a proper fit of the s-values for Pu-240. This means two separate libraries had to be made.

Reactor Engineering Development

Pu Optimization Studies

The thermal hydraulics design code, REPP, has been updated by the inclusion of several new options to make this code more general in its applications and to increase its utility. These additions include the following:

1. Flow channel preheating section option--an input specifying an entrance length in the coolant channel free of fuel may be provided.
2. Flow channel post heating section option--an input specifying an exit length in the coolant channel free of fuel may be provided.
3. Maximum fuel temperature option--an upper limiting value may be placed on the hot channel fuel element centerline temperature.
4. Iteration control option--allows specification of a maximum number of iterations for each individual case.
5. Constant coolant flow rate option--the coolant flow rate supplied in the input data may be held constant for each case.
6. Void fraction iteration option--allows specification of a given number of variations on a particular case in which the only variable altered is the void fraction (in the context used here void fraction = 1 minus fuel fraction). The ability subsequently to vary that base case is not affected.

The survey code, FULCYC, has been updated by the inclusion of the revised design code, REPP.

Fueling Vehicle Cooling

The FERTF 8-rod fuel element has unusual cooling problems during charge-discharge because of the protective sleeve surrounding the fuel

and the required low temperatures during the handling of defected elements. This protective sleeve tends to reduce air flow through the element during normal air cooling and shields the element from the emergency water cooling system. Therefore, an analysis was performed to determine the adequacy of the normal air cooling systems. This analysis was based on an 8-rod element with a maximum specific power of 0.23 kw/ft. The results of this analysis is as follows:

1. Air Cooling

It was found that the normal air cooling would be adequate for cooling a non-defect 8-rod FERTF element in the fueling vehicle. Further calculations were performed to develop information which would allow the estimation of maximum fuel temperatures from measurements of bulk outlet temperature. These calculations were performed for fuel elements with and without the basket tube liner (inner sleeve) installed and with the assembly enclosed by the discharge cask shroud. Two methods were used. In the first method, fractions of total air flow passing outside the basket tube between the basket tube and the fuel ring and inside the fuel ring were calculated for various total air flows. From these flow splits, maximum rod temperatures were calculated by assuming that the rate of heat flow out of a fuel rod was uniform around its circumference. In general, these calculations indicated that the highest temperature of a fuel rod would be inside the rod ring. Realistically, such a temperature imbalance around the rod circumference would tend to decrease the heat flux at the inner surface and drive more heat to the outer surface. Under some conditions, the calculated inner surface temperatures were higher than those which would exist if there were no cooling at the inside of the rod ring and all heat were being driven to the outside. Consequently, a second method of calculation was used to determine maximum fuel rod temperatures in which all heat was assumed to be driven to the outside. The results of these calculations were presented as plots of maximum surface temperature versus bulk outlet temperature.

2. Water Cooling of a Defected Element

For water cooling, a flow rate of 0.5 gpm inside the basket tube should prevent fuel temperatures from exceeding 212°F if the water were spread uniformly over the surfaces of fuel rods, basket tube and center rod. (This assumes that water flow is initiated before fuel begins to heat up after removal from the reactor). However, at this flow rate, channeling of the flow and non-uniform distribution would probably exist. Some experiments to determine the flow rates which will insure that water is flowing on all fuel rod surfaces will be needed to guarantee that defected fuel temperatures can be held

below 212°F. At powers up to the 0.23 kw/ft total flows adequate to provide water flow over all fuel rod surfaces should be more than adequate to hold rod temperatures below 212°F.

3. Emergency Water Cooling of Non-defected Element

If through loss of air cooling, a non-defected element were allowed to reach high temperatures before water cooling were initiated, water running down the inside of the basket tube would flash to steam and tend to divert the incoming flow. An accurate calculation of thermal and hydraulic behavior for such a case was not possible. However, estimates of maximum fuel temperatures following loss of air cooling were made by assuming that the present emergency water cooling system was used to spray water on the outside of the basket tube. In such a case, heat transfer from the fuel rods to the basket tube would be primarily by radiation. Without a basket tube liner, maximum fuel rod temperatures of 1500 to 1700°F were estimated. These temperatures would probably cause fuel rods to rupture, but would not produce any core or cladding melting. With a basket tube liner installed, the thermal resistance between the liner and the basket tube would cause higher fuel temperatures for this method of emergency cooling. The temperatures would depend on the closeness of the fit between the basket tube and liner, but should be below the melting point of core or cladding materials.

Rupture Loop Particle Separator

A transparent plastic section was attached to the downstream end of the swirler. Several tests were then made by injecting dye and UO_2 particles into the flow stream and photographing the paths of these materials. The swirl angle was found to approximate the theoretical angle predicted (26° vs. 29°). Tests were then conducted injecting fuel particles into the flow stream upstream from the swirler while taking movies at the clear plastic section. Examination of the movies indicated: (1) the majority of the particles travel along the wall; (2) the particles travel in very well defined paths rather than randomly; and (3) the large particles seem to tumble along the wall. The results of these tests indicate that the particle separator's low efficiency is probably due to the collection ring configuration.

Clear plastic collector rings are being fabricated. These rings will be attached to the plastic pipe and movies will be taken of the collector ring section while particles are being injected into the flow stream. The results of these tests will be used to modify the collector ring configuration to increase the efficiency of the separator.

Fuel Element Testing

Testing of the basket type fuel element for the FERTF continued

during the month. The loop was operated for 486 hours at 70 gpm, 495 F, and 1050 psig. Total exposure of element at test conditions to date is 1564 hours.

Periodic inspections of both the pressure tube and fuel element were made during the month. Examinations of the pressure tube with a boroscope revealed no serious fretting; disassembly and examination of the fuel element was performed by representatives of Fuel Design and Evaluation. Several photographs were made of the lower fretting marks in the pressure tube. Prints are available for inspection.

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 is composed of determining a hydrogen limit on unirradiated tube specimens of different flaw length, and crack propagation tests at various temperatures.

Room temperature fracture behavior of slotted irradiated and un-irradiated PRTR pressure tubes is established so that current interest is concerned with temperature effects. The burst strength of PRTR pressure tubes, before irradiation, was found to decrease linearly 10 psi/°C from a room temperature strength of 35,000 psi for tubes containing a 1-1/2 inch long slot through 80% of the wall thickness. An observed decrease in burst stress of 104 psi/°C starting with a room temperature burst stress of 66,000 psi for PRTR tubes containing 3/4 inch slots appeared high. A test of 150°C on an annealed PRTR tube section containing a 3/4 inch long slot confirmed the temperature coefficient of -104 psi/°C. This behavior may be the result of the combined effect of decreasing yielding strength and increasing fracture toughness with temperature.

Electron microscope fractographs were made from replicas of the fracture surface of PRTR tubes containing 30, 150, and 275 ppm hydrogen. The ratios of ductile (dimpled) to inter-and trans-granular cleavage for the three specimens were similar. This information is being used to determine how the combined affect of irradiation and absorbed hydrogen may effect the fracture toughness of pressure tubes.

Test Reactor Operation

Operating Experience

Pertinent data for the month are as follows:

1. Production. 539.11 MWd
2. Hours Critical. 277.9

- 3. Critical Efficiency 38.6%
- 4. Total Experimental Time Efficiency 58.4%
- 5. D₂O Losses
 - a. Indicated Stack Loss (6-26 to 7-25) 715 lbs.
 - b. Physical Inventory (6-24 to 7-23) 699 lbs.
- 6. Helium Loss 114,938 scf

The Plutonium Recycle Test Reactor operated at a maximum power level of 66 MW this month. This power corresponds to a maximum specific rod power of 20.0 kW/ft for fuel elements in Ring One. Total production since the start of the batch core experiment is 1,901 MWd or 9.5% of the goal exposure. The 55 fuel element core has accumulated 1,621 MWd.

PRTR operation was interrupted by 12 shutdowns. Four were scrams caused by pressurizer pressure trips, two were caused by flow monitor trips, one was a scheduled outage for preventative maintenance work, and five were intentional shutdowns to correct operating conditions. A brief description of the outages occurring during this report period follows:

<u>Date</u>	<u>Reason for Shutdown</u>
June 26	Scrammed by flow monitor 1956, high flow. Improved crud conditions in primary system resulted in a large flow increase through the 5-micron crud filter. The outage was extended to perform fuel rod removals for FERTF Test #22 and PRTR Test #136-2 and to work on pressurizer pressure control system. The outage required 23.6 hours.
June 27	Scrammed by pressurizer very high pressure. The outage was extended to work on the pressurizer pressure control valves and repaired valve leakthrough prior to startup. The outage required 56.2 hours.
June 29	Scrammed by pressurizer low pressure. Adjusted proportional band on P-13 stabilizing the pressurizer level control which smoothed out the pressure control. The outage required 3.1 hours.
June 30	Scrammed by spurious log reactor period. The outage required 0.9 hours.
July 2	Shut down for 9.6 hours to correct cause of high collection rates in the D ₂ O recovery system. Replaced eight leaking nozzle caps.

<u>Date</u>	<u>Reason for Shutdown</u>
July 3	Shut down for 8.3 hours to correct cause of rapid level decrease in primary heavy water storage tank. Replaced eight leaking nozzle caps.
July 7	Scrammed by pressurizer very high pressure. Stayed subcritical to run Power Test #6, Part B, and to work on pressurizer pressure control system. Found dirt in the level control system which was upsetting the pressure control. The outage required 104.1 hours.
July 11	Scrammed by flow monitor 1956, high flow. Improved crud conditions caused a large increase in flow through the 5-micron crud filter. The outage required 1.0 hours.
July 12	Scrammed by pressurizer high pressure. Extended the outage for 44.2 hours to resolve the pressurizer pressure control problem. Found the valve trim broken on the pressurizer high capacity helium supply valve.
July 14	Shut down for 0.2 hours to inject boron.
July 16	Shut down for 188.2 hours as scheduled to complete programmed repairs, tests and preventative maintenance. Primary pump #1 was replaced because of seal leakage. Replaced aluminum fuel transfer carriage with a carrier made of stainless steel.
July 24	The shutdown valve opened on a short reactor period. Reactor was then manually scrammed. The outage required 2.7 hours.

As shown above, the reactor experienced three high pressure scrams and one low pressure scram during the month. Following each scram, the pressure control system was examined closely to find and correct the cause of the outage, and in each case sufficient items were found and corrected prior to startup to indicate that the pressure control problem was solved. After the fourth scram on July 12, the high capacity pressure supply valve, H-75, was disassembled and found to have a broken valve trim. The trim was broken below the seating surface so that the valve made a tight shutoff; however, the tapered throttling plug was broken and a piece, about 3/4 inch long and 1/4 inch in diameter, was missing. The broken plug destroyed the throttling capability of the pressure control valve. The full flow capacity of the valve was present with a minimum of valve travel. Pressure control has been normal since the broken trim was replaced.

During the monthly emergency diesel generator load test, the primary pumps did not re-start automatically and the instrument M-G set did not transfer as it should to 384 power and then back to the diesel emergency power. The pump start problem was traced to a broken wire on the 2400 V switchgear door for primary pump #1. With this wire broken, the relays would not set up to allow

either primary pump #1 or #2 to start automatically. (This control wiring has been previously identified as weak due to old age, and a replacement program is in progress.)

No cause has been found for the failure of the instrument M-G set to transfer. This failure to transfer has occurred previously on monthly diesel load tests. Diagnosis has not been successful because the trouble will not reproduce itself.

The water chillers experienced five failures this month. Vibration of the compressor discharge piping is causing fatigue failure of the oil separator tubing. Snubbers have been installed (NPC-446) to reduce the vibration to a minimum.

The testing of the gas loop blowers (a portion of the lay-away program for the gas loop) has become increasingly difficult. Gas blowers 1 and 3 finally would not operate. Therefore, all blowers were removed from service. Immobilizing flanges were fitted on the inlet flanges to take the weight off the special bearing and eliminate the need for periodic testing. A dry atmosphere was provided.

The ion exchange resin was removed from the FERTF piping. Two new ion exchangers charged with LiOH resin were installed, and the FERTF was reconnected to process channel 1550. Oxygen concentration, previously a problem, has remained within limits since the FERTF has been in service with the fresh ion exchangers.

The amount of reddish-violet material visible on the fuel elements continued to decrease during the month, indicating that crud levels in the primary system remained at low levels. New 10- and 40-micron pore size filters were installed in place of the 5-micron filters which had been used to date. After three days of full power operation, the new filters showed little evidence of plugging.

Four process tubes were examined. There were no significant changes observed since the previous examination. Eleven irradiated fuel elements were inspected in the storage basin. All were in satisfactory condition. Nine irradiated fuel rods were shipped to the Radiometallurgy laboratory in support of PRTR Test 136 and FERTF Test 22.

Process Technology

The fuel rod being irradiated at 19.7 kW/ft under Supplement 2 of PRTR Test 136 was subjected to a maximum of 20.0 kW/ft during a reactor startup and rise to power on June 26, 1967. Metallurgical examination of this fuel rod revealed that the onset of fuel melting had been reached (8% molten radius). A similar power fuel rod from another fuel element was removed under FERTF Test 22 and its examination revealed a 30% molten radius (a power 5% above the onset of melting). As a result of these two examinations, the allowable specific rod power was conservatively reduced. At month end, with the FERTF in service, the limit was 17.1 kW/ft on a "maximum tube" basis rather than the previously used "average of

the six central tubes."

An investigative study of the molten fuel event and a re-evaluation of the long term program were also begun. The significant accomplishments during the month were:

- . The removal of six additional irradiated fuel rods for metallographic examination;
- . The installation of new fuel rods on three of the central fuel elements and the positioning of these rods in the peak power positions for additional irradiation and examination phases;
- . The installation of two special fuel element bands on each of three central elements for investigation of real power differences between these process channels;
- . The completion of a series of axial peaking factor measurements during a recent cold startup which reaffirmed the degree of conservatism in the procedural controls;
- . The determination that the tube data reduction system was working improperly during the reactor startup and rise to power on June 26. (During the startup and rise to power, fuel rod specific powers reached 20.0 kW/ft. When this was recognized, testing and diagnosis were conducted to correct the problem. The system is now functioning properly.)

The startup and the shutdown phases of Batch Core Power Test No. 6 (Transient Fission Product Poisoning) were conducted during the month. Preliminary analysis of the data indicates that the poisoning associated with the growth of equilibrium xenon is in good agreement with calculated results. The magnitude of the xenon poisoning transient following shutdown from equilibrium power operation was approximately half that expected. Further investigation of this apparent anomaly is in progress and a test supplement for another measurement of the initial 24 hours of the shutdown transient has been prepared.

The two fission gas pressure measuring fuel elements continue to perform satisfactorily. Thirty pressure and temperature measurements were made on each of the four test rods. The maximum temperature and pressure observed were 529°F and 58.2 psig in rod 8 of fuel element 6520 located in process channel 1548. Fuel element 6520 was discharged to replace a leaking nozzle cap gasket. While the process tube was empty, it was inspected and found to be in satisfactory condition. This was the first time an irradiated fission gas pressure measuring element had been discharged.

Coupon samples were removed from the primary system, examined for corrosion and film deposition and returned to the system. Measurements from these samples are still being evaluated.

Coupon samples were reinstalled in the out-of-reactor test section of the Fuel Element Rupture Test Facility. These samples will provide long term corrosion and film sureillance.

Aluminum corrosion probes in the moderator and reflector cooling systems, and carbon steel probes in the top and bottom shield cooling systems and in the core blanket gas system all showed corrosion rates of less than 0.01 mils per month. Carbon steel probes in the biological and thermal shield cooling systems showed rates of 0.13 to 0.24 mils per month in these systems which are cooled with process water.

The HX-5 tube bundle inspection plate was removed and the tube bundle inspected for mechanical damage. Results were satisfactory.

Initial testing of the pneumatic accessory was begun. The exhausted irradiated air has not been a problem.

Improvement Work Status

Work Physically Completed.

NPC-427 - Fuel Transfer System Improvements. The carriage used to carry fuel elements between the containment vessel and the storage basin was originally fabricated from aluminum. It proved to be structurally weak and was also damaged by corrosion. It has been replaced by a stainless steel carriage.

NPC-372 - Moderator Filter Shielding. A sideleg particle filter was installed on the moderator system after the FERTF incident. Shielding design for this filter was delayed to await moderator system experience with the reactor at full power. A one-inch-thick lead shield has now been added.

DC-300 - Coarse Particle Filter. A 40-micron filter and particle trap have been installed in an empty primary process tube in addition to 10-micron filters installed in two other tubes. The particle trap will catch larger particles that might not cling to the outside of the filter and ensure that any such particles will not remain loose in the primary system.

NPC-347 - Chemical Addition Pumps. The addition of chemicals to the boiler feedwater has been made more flexible by re-arranging the piping system on the chemical addition pumps.

DC-309 - H-78 Valve Removal. H-78 is a relief valve designed to relieve helium from the pressurizer to IP helium storage for small overpressures thereby saving helium. (Larger overpressure conditions relieve to the cell via primary system relief valves.) The H-78 valve has been a frequent source of maintenance problems and a recent review showed that the valve actually works on a pressure differential between the two systems. Since the downstream pressure (IP storage) varies widely in the course of normal operation, H-78 is not capable

of relieving at a predetermined upstream pressure. Therefore, it has been blocked closed and will be removed when suitable blanks are obtained.

EBWR Demonstration Program

Irradiation Testing of EBWR Prototype Fuel Rods.

The last two of the original group of 32 capsules are being examined. The Zircaloy-clad (1.07 cm OD) capsules contain vibrationally compacted, pneumatically impacted UO_2 -1.5 wt% PuO_2 . Irradiation conditions relating to these two capsules are summarized in the following table:

IRRADIATION CONDITIONS FOR CAPSULES GEH-14-523 & 524

<u>Capsule Number</u>	<u>Effective Full Power Days</u>	<u>Flux, $\times 10^{14}$</u>	<u>Rod Power, kW/ft</u>	<u>Exposure MWd/tonne</u>	<u>nvt</u>
GEH-14-523	899	0.94	16.7	27,600	7.0×10^{20}
GEH-14-524	926	0.84	14.9	26,300	6.7×10^{20}

Each capsule has a void volume of 0.9-1.0 ml and contains 51.6-51.8 g of mixed-oxide fuel. Fuel and gas samples are being analyzed to establish actual burnup and gas-release values. Gas release from these capsules was 12.1 ml for capsule GEH-14-523 and 7.4 ml for capsule GEH-14-524.

Isotopic data from three other capsules indicate that for burnups of 12,560, 16,930, and 18,300 MWd/tonne, the $^{241}\text{Am}/^{243}\text{Am}$ ratios are 3.00-3.03, 1.83-1.88, and 0.86-0.88, respectively.

Examination of irradiated production-run rods is continuing. Ceramographic examination of the rods which attained an estimated burnup of 27,500 MWd/tonne was initiated. Fission gas and fuel samples from these rods are being analyzed.

NUCLEAR SAFETY

Containment Systems Experiment

Containment Leakage Studies - Task A

Effect of Temperature Sampling Errors in Leakage Rate Measurements.

A serious source of error in determining the leakage rate of containment vessels is use of erroneous average containment air temperatures. Such errors may result from inadequate sampling of the air volume. During the leakage rate tests on the CSE containment vessel, air temperatures were measured by 19 platinum resistance temperature detectors (RTD's) distributed to provide one RTD for each 1600 ft³ of containment vessel volume. Leakage rate calculations by the absolute pressure-temperature method were based on changes of the volume weighted average temperature of all 19 RTD's.

Leakage rates were calculated to show the effect on absolute method calculations if the temperature sensors were used individually and if disadvantageous locations are used. An example of some of this data at a moderate leakage rate is shown in the following table:

CONTAINMENT LEAKAGE BY THE ABSOLUTE METHOD (24 HOURS)

Li = leakage calculated using individual sensor
L = leakage from volume weighted Avg. of all 19 RTD's

<u>POINT NO. LOCATION</u>	<u>Δ TEMP. (° F)</u>	<u>Li (%/day)</u>	<u>Li/L</u>
1.	1.46	.245	0.85
2.	0.83	.363	1.25
3.	1.50	.238	0.82
4. Wetwell	0.46 *	.432	1.49
5. Access way	0.70 *	.387	1.33
7.	1.28	.279	0.96
8.	1.38	.260	0.90
9.	1.56	.226	0.78
11. Bilge	0.36 *	.450	1.55
13.	1.40	.256	0.88
14.	1.34	.267	0.92
15.	1.54	.230	0.79
17.	1.44	.249	0.86
18.	1.16	.301	1.03

POINT NO. LOCATION	Δ TEMP. ($^{\circ}$ F)	Li (%/day)	Li/L
25.	1.48	.241	0.83
26.	1.50	.236	0.82
28.	1.43	.249	0.86
29.	1.50	.238	0.82
30. Bilge	0.47 [*]	.429	1.48
Average	1.20	.290%	1.00

* This and other data indicates that reduced number of individual sensor can give a fairly adequate sample of containment air temperature as long as obviously disadvantageous combinations are avoided, such as using all bilge and wetwell sensors for an average..

Fission Product Transport Studies - Task B

Aerosol Transport and Behavior in the Drywell Vessel

A second aerosol transport test was performed in the CSE drywell vessel. The objective of this test was to check equipment and methods and to train operators for the forthcoming full scale CSE experiments. The test duplicated the prior drywell test except that ruthenium and zircaloy-UO₂ fumes were added to the aerosol. The painted walls of the vessel had been aged by previous steam exposure for this second test.

Preliminary results show the gas phase concentration half lives for iodine, cesium, ruthenium and uranium was 12 + 1 minute. In the previous test, the half lives for iodine and cesium were 8 minutes. The longer half life is at least partly attributable to the lower steam condensation rate in the latter test. The half life predicted by the ADF mathematical model was 11.5 minutes if the bulk gas velocity near the walls was 60 ft/min. Gas velocity measurements were made but interpretation awaits anemometer calibration under test conditions. Significant test conditions are listed below:

Vessel Properties	height	7.6 m
	diameter	3.3 m
	volume	65 m ³
	surface area	100 m ²
	coating	Phenoline 302
	insulation	none
Previous steam exposure	33 hrs at 250 ^o D, 42 hrs at	

of painted surfaces.	212°F Continuous steam feed to replace loss by condensation.
Afterheat	
Temperature	121 + 1 °C, isothermal
Pressure	49 + 1 psia, isobaric
Steam to Air Ratio	1.6 mo./mol
Steam Condensation Rate	2700 + 100 g/min
Initial Aerosol Conc. (calculated)	
Iodine	0.9 mg/m ³
Cesium	5 mg/m ³
Ruthenium	0.04 mg/m ³
Zr-UO ₂	~10 mg/m ³
Furnace sweep gas, air, 2 CFM	
Release duration, 10 minutes	

This was the last intermediate scale transport test, and future tests will use the 30,000 ft³ main CSE vessel.

CSE Aerosol Characterization

The conifuge is to be used as a method of determining the settling velocities of all components in CSE aerosols as injected.

Cone liners are being developed so that the entire deposited sample can be used for analysis.

To date, paper and aluminum foil liners, have been tried with poor results. The liners apparently fluttered at the top of the cone or pulled loose due to the varying centrifugal force down the cone.

Strips of masking tape cut to size on a template are now being used as a cone liner with good results. Tests will now resume on developing a method of tagging polystyrene latex spheres for use as an internal standard.

Small Scale Aerosol Tests

Two runs were made in the Aerosol Development Facility (ADF) to enable comparison of aerosol analysis by CSE techniques and ORNL techniques. BNW and visiting ORNL personnel cooperated closely to analyze aerosol samples from a typical ADF run. Two runs using

Zr- UO_2 fumes, I_2 , Cs, and Ru were made in steam and air at 80°C . The ORNL device² included two-six Maypack clusters, one of which was loaded with carbon filmed membrane filter for electron microscope work, a fibrous filter analyser, and a composite diffusion tube.

The samples taken by the ORNL employer are smaller than the typical ADF Maypack samples and they are broken in to more fragments for counting. This, along with lower than expected release fractions during the aerosol generation, and sample flow problems caused by the atmosphere pressure in the ADF run, gave low counting levels.

These shortcomings were corrected and a second run was made. The results from this run are being analyzed; and it appears that satisfactory sample activities were obtained. The levels were ample for useful analysis of the ADF samples in both runs.

CONTAINMENT SYSTEMS EXPERIMENT-R. L. Dillon

Decontamination

The facility used to conduct experiments on the decontamination of Phenoline 302 painted surfaces was modified and calibrated and is now ready to continue experimental operation. Calibration of this apparatus refers to pressure versus flow characteristics of the spray jets used to clean contaminated surfaces.

Coolant Blowdown Studies - Task C

Blowdown Experiments

One major portion of the CSE program involves large scale coolant blowdown experiments which have the basic purpose of investigating the dynamic effects of large ruptures in high temperature reactor cooling systems. Data on the transient pressures, temperatures and forces imposed on the containment vessels, reactor vessel and internal components will be measured for comparison with values predicted by analytical models. This information will be used to establish the validity and range of applicability of analytical methods used in safeguards analyses of nuclear generating stations.

Two hot blowdown tests were made with the reactor simulator vessel. In the first test, a full vessel of water at 500°F and 665 psig (saturation pressure) was blown down through a 3,438" I.D. orifice at the end of a bottom blowdown nozzle. In the second test, water at the same initial conditions was blown down through the full open pipe of 6.8" I.D. The purpose of these tests was to check the blowdown predictions made by various authors, and to continue the development of instrumentation techniques.

Liquid level probe and total weight load cell measurements

were taken successfully on both runs. There is some problem with zero and initial reading drift during heat up for a hot run. Despite this drift, the liquid level decrease in the tank and the weight loss could be followed quite well for these high temperature runs.

The results of the blowdowns do not agree with either homogeneous or annular equilibrium two-phase critical flow model predictions. The experimental blowdown rates were roughly 60% of those predicted. It was noted that analytical models proposed by BNW⁽¹⁾ and General Electric predict discharge rates low by about this same factor. It may be further noted that this factor of 60% is similar to the nozzle coefficients of 0.698 to 0.70 needed to make the ambient temperature - nitrogen dome water blowdown results agree with adiabatic theory. The pressure gages and temperature gages, both in the vessel and in the discharge pipe, do not agree among themselves or with steam table values better than 10%. The results will require further interpretation to explain all the data.

Radioactive Waste Solidification (A. M. Platt)

Pot Calcination

Process Technology.

B-Cell Run PC-5 will be a pot calcination run with PW-1 as feed. The actual feed will differ from nominal PW-1 by having somewhat higher sodium, aluminum and sulfate concentrations. Feed solution was simulated in the laboratory and its stability and boiling characteristics examined. Foaming is a recurring problem in the pot-cal process and previous laboratory tests have proven inadequate for predicting potential foaming conditions. Simulated Run PC-5 feed was foam-tested in a new long tube configuration. The only significant foaming observed was apparently due to superheating which was dissipated by large surges of foam lasting 20-30 seconds.

Radioactive Demonstration.

Pot calcination run 4 (PC-4) was successfully completed using a modified PW-1 feed composition. Excessive sodium in the Purex IWW used for feed make-up required the addition of an equivalent amount of sulfate to prevent formation of sodium and cesium oxides in the calcines. Thus the sodium and sulfate concentrations were 0.47 and 0.26 respectively.

A total of 650,000 curies, including 47,000 curies of radio-ruthenium and a strontium-90 spike of 30,000 curies was fed to the

(1) B. M. Johnson, "Mathematical Models of Pressure-Temperature Transients", BNWL-233, May 1966

12-inch pot in 42 hours at an average feed rate of 26 liters/hr. The high feed rate was primarily due to some concentrating of the feed in the evaporator since a C-mode equipment arrangement was used. This consisted of feeding the pot from the boiling waste evaporator and returning all of the pot condensate to the evaporator for recycle with the feed. The evaporator was continuously fed from a batch feed tank.

During the first few hours of feeding the pot, a severe foaming condition was encountered and approximately 20 liters of concentrated feed was regurgitated from the calciner into the safety relief seal pot. After feeding water to the pot for approximately one hour, the run was successfully continued. A reduced feed rate was used to allow more freeboard for foaming and no further foaming was observed.

Drying and cooling of the pot contents after the feed was turned off required 33 hours to give an average processing rate of 14.4 liters/hour. 166 Kg of calcine was netted with an internal heat generation rate of 2700 watts. This produced a pot centerline temperature of 477°C and a centerline to wall ΔT of 310°C with the pot suspended in 38°C air.

One of the principal features of the mode C equipment setup is recycle of the pot condensate with incoming feed to the pot. Thus, a constituent which is volatilized from the pot will increase to some steady state concentration in the evaporator during a series of similar runs. An objective of this run was to observe the recycle effect on ruthenium. However, only five percent of the ruthenium in the total feed to the pot was volatilized as indicated by data from stream samples of the pot condensate. Final samples of the feed remaining at the end of the run revealed no increase in ruthenium and confirmed the low volatility. In earlier pot runs with PW-2 waste, 30 percent of the ruthenium was volatilized.

Additional removal of ruthenium from the gas stream was sought by adding sodium hydroxide to the tower of the acid fractionator. Preliminary data indicate that the ruthenium DF may have improved by a factor of 10 over previous runs. A comparison is planned by not adding caustic to the fractionator during the next run, PC-5.

All equipment operation during the run was satisfactory except for semi-continuous plugging of the waste evaporator specific gravity dip tube.

Phosphate Glass Solidification

Process Technology. The rough draft was completed on the terminal report (Part V) on the laboratory studies associated with the Waste Solidification program.

At the request of Waste Solidification Engineering, a sample of glass from run CPG-7 in the 324 Building phosphate glass equip-

ment was measured for thermal expansion. Below the glass transition temperature, $\alpha = 1.4 \times 10^{-5}$; above the transition temperature, it was 9.9×10^{-5} . The glass transition temperature was 380°C and the dilatometric softening temperature was 415°C .

Spray Solidification

Process Technology. The corrosion rate of 304 L stainless steel was determined in calcined PW-1 solution containing NaPO_3 at five concentration levels (Meq/P from 1.5 to 2.3). The corrosion rates (liquid phase) ranged from four to 23 mils/mo. with no preferential attack at the liquid-vapor interface. These data indicate the feasibility of by-passing the platinum melter and melting directly in the storage container when utilizing these feed compositions. The above melts were less corrosive to 310 stainless steel (factor of about five) than 304L. PW-1 calcines containing Colemanite at three concentration levels (510, 610, and 710 g/l PW-1) had melting points ranging from 800 to 840°C . Corrosion rates of 304L in these melts at 850°C ranged from 100 to 235 mils/month.

Engineering Development. Attempts are being made to improve the reproducibility (presently on the order of $\pm 10\%$) of the drop size measuring technique. Possible sources of error which are being checked include: insensitive flowmeters, drop coalescence, and random sampling error. Although the absolute values of our drop size measurements did not agree as closely as would be desired with those made by Delavan Company, the Delavan measurements confirmed the superior performance of the internal mix nozzle compared to an external mix nozzle.

An eroded internal mix nozzle was compared with a new nozzle and no difference in droplet size could be seen.

Product Evaluation

Leach Rate Studies - Phosphate Glass. Leach rates for two samples of phosphate glass (PW-2) product taken from different locations in the same collection pot (WSEP Run B) were compared. One sample was partially devitrified and the other completely glassy. Devitrification of the one sample was due to its being held at elevated temperature longer than the glassy sample. The leach rate of the devitrified sample was about two orders of magnitude higher than that of the glassy sample; it was about the same as the leach rate for microcrystalline spray solidifier PW-1 product.

Leach Test - Spray Solidification. A series of leach tests were made with grab samples taken during an EDL spray solidifier PW-1 run (Run DSC-1A) and with samples taken from the pot on completion of the run. Results of the tests indicated that calculated leach rates are relatively independent of both frequency at which leach water is sampled and changed (over a range of once a day to once every two weeks), and leachant volume (over a range of 100 to 500 ml of leachant with samples having about 15 sq. cm. of surface area).

In three separate comparisons, as-received grab samples were significantly more soluble than pot samples collected at a corresponding time during the run. These results are contrary to what was expected and more comparisons of this type will be made.

Heat Generation Rates. Heat evolution rates of 2.6 and 1.1 Kilowatts were measured with the in-cell calorimeter for pots from runs PC-4 and PC-2 respectively. Agreement was within 10 to 20 percent, respectively, of values predicted from material balance and decay curves.

Solids Storage Engineering Testing

The requirements for installing at least two pods in B-Cell, prior to achieving beneficial occupancy of A-Cell, have been analyzed. The results indicate that two pods can be installed in Buvice B-14 without removal of process jumpers, but that some additional handling fixtures will be required over and above that needed for A-Cell. Pod installation can begin as early as 9-1-67, following fixture design and fabrication during August.

Project BAP-016, Solids Storage Engineering Test Facility, is underway with the start of wall coring required for installation of the in-the-wall intercubical piping. Installation of the wall liner is delayed, pending completion of electrical details for new incell lighting at the upper level.

The most critical scheduling item, procurement of the second shielding window, appears on schedule, following placement of the purchase order and commitment by the vendor to deliver the frame and window on the required dates. These commitments, if kept, place the proposed A-Cell startup date of January, 1968, in favorable light.

Fission Product Aerosol Containment

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

The effect of much finer spray droplets₃ on removal rate of methyl iodide was determined using the 3.6 m³ spray chamber. A 4.8 wt % hydrazine - 5 wt % ammonium hydroxide solution was sprayed through an air-steam atmosphere containing 0.1 mg of methyl iodide per cubic meter at a rate of 0.108 gpm. A Bete Pin-Jet nozzle producing a spray with a median mass diameter of 140 microns was used. Initial temperature in the chamber was 88°C which decayed to 43°C during the 6 hour spray run. Approximately 58.7% of the recovered activity was contained in the spray solution at the end of this time. The removal half-time was 4.2 hours under these conditions compared to a removal half-time of 21.2 hours for an equivalent flow of water assuming a partition coefficient of 5. The true partition coefficient for methyl iodide probably varies from approximately 0.2 to 90°C to 1.0 at 45°C.

For a given throughput of liquid, the 5% hydrazine delivered as a fine spray gave significantly better removal rates than were

achieved in earlier runs with coarser sprays. The conditions in this run were such that the hydrazine-methyl iodide reaction was effective, even though the dwell time in the spray chamber was short.

Progress was made toward readying the Aerosol Development Facility vessel for higher pressure and higher temperature demonstration runs.

Effort continued on the development of a suitable model for the washout of CH_3I by reactive liquid sprayed into a chamber. The liquid phase reaction rate required by double the removal rate of methyl iodide (expected from only physical absorption) was calculated on the basis of the penetration theory for a first order irreversible reaction. The required reaction velocity depends upon the reaction time available during the passage of the droplets, hence is a strong function of the drop size. When applied to a wall wetted with the solution, the relationship shows that removal rate will be a marked function of film thickness. The initial analysis showed that the reaction rate determined experimentally for 5% hydrazine and methyl iodide was not rapid enough, for most experimental conditions employed, to double the washout rate when droplets fell nine feet, the fall height of droplets in the spray chamber used. Smaller droplets and higher hydrazine concentrations would appear to significantly enhance the removal rate. These theoretical concentrations are reasonably consistent with the experimental observations to date. As earlier indicated, drops with a median size of 140μ do show a significant removal rate over that predicted for water.

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

The reaction rate constants for the liquid phase reaction of hydrazine and three other compounds with methyl iodide were determined using earlier developed methods. Results are shown in the table. Rate constants are reported both as pseudo-first order rate constants and as second order reaction rate constants.

Reaction Rate Constants

<u>Description of Solution</u>	<u>Observed Half-Life (min)</u>	<u>Pseudo-First Order Rate Constant (min^{-1})</u>	<u>Second Order Rate Constant ($\text{M}^{-1}\text{sec}^{-1}$)</u>
0.64 wt. % N_2H_4 in 0.0235 N HCl	33.7	2.1×10^{-2}	1.7×10^{-3}
0.64 wt. % N_2H_4 in 0.0255 N NaOH	1.7	0.41	3.4×10^{-2}
0.9 N $\text{Na}_2\text{S}_2\text{O}_3$	4.2	0.165	3.1×10^{-2}
0.007 N $\text{Na}_2\text{S}_2\text{O}_3$	57	0.012	2.6×10^{-2}

Description of Solution	Observed Half-Life (min)	Pseudo-First Order Rate Constant (min^{-1})	Second Order Rate Constant ($\text{M}^{-1}\text{sec}^{-1}$)
0.09 N KIO_3	39-270*	0.023	2×10^{-2}
0.0504 N NaOH	4.4×10^3	1.6×10^{-4}	5.3×10^{-5}

Basic solution of hydrazine react much faster than do acidic solutions as indicated. Sodium thiosulfate is included as a reference, since solutions of sodium thiosulfate are being advocated for sprays in reactor containment. Reaction rate of the thiosulfate with methyl iodide were comparable to the hydrazine-methyl iodide rate.

A principal effort was made to further improve and verify the method for measuring the homogeneous reaction rates. The method continues to give valuable data and will be used in screening prospective compounds. Hydrazine and sodium thiosulfate rates will be used as reference comparison rates in the search for improved scavengers of methyl iodide.

During the reporting period, a paper, "The Reactions of Hydrazine and Iodomethane. The Removal of Iodomethane from a Gas Phase in Contact with Alkaline Hydrazine Solution," U. S. Atomic Energy Commission Report BNWL-SA-1035, was presented at the 22nd Northwest Regional American Chemical Society Meeting, Richland, Washington. Also, the preparation of a manuscript, "The Partition Coefficient of Methyl Iodide," was begun.

Disposal of Reactor Off-Gas into Soil Systems (L. G. King)

In modeling the field situation for gas disposal at the NRTS site, it is important to know the elevation of the surface of the basalt. Data from the twelve wells drilled to date are being used together with seismic data to determine the basalt depths. Some differences in depths exist between the two sets of data; the seismic results will be reanalyzed with the aid of a computer program in an attempt to resolve these differences. The well data are being fitted with polynomials through the use of the GENORO program. Ground surface elevations have been requested from Idaho Falls. These elevations are needed in order to convert the depths of basalt below ground surface into elevations of the top of the basalt.

Waste Solidification Condensate Treatment (B. W. Mercer)

Laboratory studies are being conducted to determine the effectiveness

* Considerable scatter of data, so that an upper and lower limit of half-life was estimated

of electrodialysis and ion exchange for decontaminating WSEP acid fractionator condensate. The condensate obtained for these studies contained 0.03 mole/liter nitric acid, 1×10^{-2} $\mu\text{C}/\text{ml}$ of Ru^{106} and 6×10^{-3} $\mu\text{C}/\text{ml}$ of Ce^{144} . Condensate was pumped through an electrodialysis stack pack at a flow rate of 1 liter/min. The stack pack contains 20 cell pairs with 220 cm^2 of membrane surface area. The current through the stack pack was 3.5 amperes and the diluate to concentrate ratio was 10:1. About half of the Ru^{106} and 93% of the nitric acid was removed from the diluate stream. The diluate was recycled through the stack pack to simulate a second stage which was limited to 0.6 ampere. Since most of the nitric acid was removed in the first stage, the resistance of the diluate is high. The overall nitric acid and Ru^{106} DFs for the two stages was 128 and 5.5 respectively. Cesium results are not available at the present time. Although ruthenium removal by electrodialysis is limited, the removal of the nitric acid facilitates final cleanup of the condensate by ion exchange. Preliminary data with an anion exchange resin column indicate that an additional ruthenium DF of 20 possible by ion exchange.

The removal of ruthenium and nitric acid by ion exchange alone is also being studied. Results with a cation resin column in series with an anion resin column show ruthenium Dfs greater than 100. The condensate was pumped through a cation resin column first to remove cationic activity and ions such as Fe^{+2} which might precipitate in the anion resin column. The anion resin column is effective for ruthenium removal until acid breaks through the column. From 24 to 33 column volumes of condensate was pumped through a strong base anion resin column to ruthenium and acid breakthrough. A weak base resin column was effective to 48 column volumes; however, the ruthenium DF slowly decreased during the latter portion of the run.

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

Results of seven drill-stem tests taken at various intervals in the Rattlesnake Hills No. 1 Well were received from Cook Testing Co., Long Beach, California. An evaluation of the data suggests that permeabilities in the test zones are generally low. Elevation of the potentiometric surfaces generally decreases with depth from +1829 feet above mean sea level for the shallowest test zone to -4691 feet below mean sea level for the deepest test zone. The ground surface elevation of the well is +2880 feet above mean sea level. The tests further indicate that a high degree of communication does not exist between the different zones and that fluids injected into formations at depth would move downward if movement should occur.

Columbia River Sedimentation Studies (D. R. Kalkwarf)

Radiochemical analysis of water and suspended-sediment samples from the Columbia River estuary continued in support of the U. S. Geological Survey activities there. Better coordination between the sampling schedule and the radionuclide-counting schedule has

minimized the delay in radiochemical analysis; 70 samples were received this month.

Plastic containers made of linear polyethylene and used by a service organization to collect reactor effluent samples for this program were tested for their capacity to retain radionuclides by absorption. Following storage of a typical sample for one day, a re-emptied container was found to retain 20% of the Sc^{46} , 6% of the Mn^{54} , 95% of the Zr^{95} - Nb^{95} and 45% of the Sb^{124} on its walls. Addition of nitric acid to the containers prior to sample collection was recommended.

Pressure Vessel Crack Monitoring

Detection of Metal Overstress by Acoustic Emission - P. H. Hutton, N. E. Ord, J. F. Dawson, D. M. Romrell

Development efforts continued on the program to utilize acoustic emission as a means of detecting flaw growth in reactor pressure piping. A system analysis study, which will consider various parameters and functional approaches to arriving at a practical monitor system, is in progress. The current technology is at a stage where such a study is feasible and desirable to guide the most efficient expenditure of future effort. The study will provide a comprehensive guideline as to complexity, reliability, and further development work required to attain monitor systems of varying degrees of refinement.

Three recent burst tests using 3" dia. A-106-B carbon steel pipe pressure vessels distinctly support two hypothesis concerning acoustic emission. There is definite evidence indicating that the readily detectable signal from deformation in this material as a crack grows (the continuous signal of varying amplitude) is generated by the attendant fracture of the oxidized surface layer. For "as-received" specimens, the continuous signal was quite evident; but in a similar specimen with the oxide layer removed by sand blasting the continuous signal could not be detected even with signal amplifications of 10^4 . There was an abundance of transient signals throughout both tests leading up to specimen failure. These tests also indicated that the continuous signal, when present, plus some of the burst or transient signals are transmitted by surface wave propagation modes. Most of the transients appear to be transmitted by the shear wave mode. This information was derived by comparing the response from recessed and surface mounted transducers, where the recessed transducer would discriminate against surface waves.

A preliminary model of a high temperature piezoelectric transducer has been assembled and tested for four hours at 610°F and three hours at 680°F with encouraging results. The transducer maintained a satisfactory level of sensitivity throughout the test and produced satisfactory response up to 500 kHz. The first models of this transducer are being fabricated using PZT-5 crystals in preparation for monitoring a burst test of a 24" dia.

x 1.7" wall A-106-B carbon steel pipe in collaboration with the Battelle-Columbus laboratories. This is with the realization that PZT-5 has a limited useful life at temperatures in the 600°F range. Subsequent models will incorporate higher temperature piezoelectric materials.

A requested summary of the acoustic emission program to date has been submitted to ORNL for the first installment of routine coverage of this topic in "Nuclear Safety", a technical progress review publication.

ASSISTANCE TO CUSTOMERS - (J. C. Fox)

Shielding

Several computer codes that have been obtained from the Radiation Shielding Information Center at Oak Ridge National Laboratory have been made operational on the local machines. The test problems for the QAD series (QAD-IV, QAD-B, QAD-P5A and QAD-V) were successfully run and the answers obtained agree with those supplied by RSIC. The CHARGE code, which computes proton, neutron, electron and gamma ray fluxes, is currently being checked out on the IBM-7090, prior to placing it on the UNIVAC-1108.

Throughout the debugging of the QAD codes, numerous problems were encountered that were attributed to the method used by the 1108 software to handle indices. This problem will be encountered in practically any case when an offsite code is being made operational on the 1108.

The QAD code is currently being used to calculate the amount of gamma ray heating in material test samples that have been irradiated in the Engineering Test Reactor.

A library of evaluated gamma ray cross section data is being constructed. This library will contain the best currently available detailed cross section data and should help alleviate at least one problem that has been continually associated with gamma ray calculations, namely basic data inaccuracy.

Fuels Support

The results of the calculations performed in support of the fuels irradiation in the EBR-II have been transmitted to the Materials Department. These calculations were made to determine parameters, such as reactivity effect and flux depressions or peakings, relative to standard EBR-II driver elements. Also, an analysis has been made of the plutonium buildup in a depleted uranium pellet that is driven by a fast flux typical of that produced within an EBR-II driver element.

A comparison between calculated and experimental heat generation rate in a uranium oxide fuel pin irradiated in the TREAT

reactor shows that the values calculated differ by about 10% from those measured in a transient experiment. Part of the discrepancy that was noted is attributed to the difference in the design of the autoclave used to contain the fuel specimen.

Flux depression factors, thermal flux profiles and plutonium isotopic concentrations have been obtained for a plutonium oxide fuel sample irradiated in the K-East Reactor. The results of these calculations will be used to determine the average flux in the fuel pin.

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