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REACTOR & MATERIALS TECHNOLOGY MONTHLY REPORT
APRIL 1965

F. W. Albaugh

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REACTOR AND MATERIALS TECHNOLOGY DEPARTMENT
MONTHLY REPORT - APRIL 1965

Compiled by Members of the
Reactor and Materials Technology Department
PACIFIC NORTHWEST LABORATORY

May 14, 1965

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REACTOR & MATERIALS TECHNOLOGY DEPARTMENT MONTHLY REPORT

APRIL 1965

TECHNICAL ACTIVITIES

A. FISSIONABLE MATERIALS - O2 PROGRAM

1. Metallic Fuel Development

Alternate Uranium Composition. Operating conditions for driver fuel elements of a single tube design and extended goal exposure require development of a fuel material more swelling resistant than the current 150 Fe - 100 Si N-fuel composition. Previous monthly reports have described evaluation tests that are in progress for materials fabricated to single tube geometry and both beta and gamma heat treated. The alloys being studied are shown in the following table with the calculated volume of second phase compound present in each. Data obtained during the past month on recrystallization and hot hardness of the 350-350-800 alloy are also shown. In the March report it was observed that the materials would be expected to fall into three groups if the volume of small second phase particles are an indication of behavior. It is seen that the hot hardness values do have this relationship and that the recrystallization and hardness recovery data obtained thus far also show a correlation. Stress cycling tests at 400 and 500 C of all the materials are being completed to 5000 cycles.

Studies are in progress to evaluate the irradiation performance of uranium base alloys in terms of composition, fuel fabrication history, corrosion behavior, and irradiation history. An irradiation test of 120 specimens of 17 uranium base alloys containing small additions of Zr, Mo, Nb, Al, Si, Fe, P, and C in various conditions of heat treatment is being conducted in NaK capsules. Calculations based on thermocouple temperature measurements and neutron flux distributions along the NaK capsule columns show that maximum uranium temperatures range from 625 C at the center of the capsule columns down to approximately 500 C at the ends. The temperature differences are a result of an unexpected peak in the reactor flux. A goal exposure of 3000-4000 Mwd/ton is planned for these tests.

Fluted Fuel Element Evaluation. The examination of an irradiated N-single tube fuel element from an ETR irradiation test was started. This fuel element is a companion piece to one which failed during its 13th cycle of irradiation. The elements had received an exposure of approximately 2500 Mwd/ton. The shape of the fuel outer surface was intended to permit some swelling without creating high tensile stress in the outer clad. This was possible because the fluted shape permitted outward deflection of the clad, with only bending strains, up to

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EVALUATION OF URANIUM BASE FUEL MATERIALS

Alloy	Composition			Volume Percent Compound	Hot Hardness Kg/mm ² *				Recrystallization Temperature**		Hardness Recovery Temperature***	
	Fe	Si	Al		300 C		400 C		Beta	Gamma	Beta	Gamma
	ppm	ppm	ppm		Beta	Gamma	Beta	Gamma				
1	150	100	-	0.62	94.6	93.1	60.3	60.8	475 C	480 C	445 C	445 C
2	250	350	-	1.81	98.5	101.7	70.8	75.1	505 C	505 C	465 C	470 C
3	250	250	250	1.69	94.8	96.9	71.7	75.0	505 C	505 C	455 C	465 C
4	350	-	800	1.74	105.0	101.2	68.8	74.6	500 C	525 C	450 C	465 C
5	350	350	800	2.99	129.0	123.5	85.9	90.6	-	-	-	-

*Average of tests run at 400, 600, and 800 pound loads, 4 hours loading, 10 mm WC ball.

**Temperature for 50% recrystallization in 20-hour anneals of 21.4% cold-worked material.

***Temperature for 50% decrease in 20-hour anneals of 21.4% cold-worked material.

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a volume expansion of 11%. The shape also imposed less clad restraint on the uranium and thereby lessened buckling forces on the inner clad.

Examination of the irradiated fuel indicates that the design performed as intended in regard to accommodating swelling; however, unexpectedly large amounts of swelling occurred in some portions of the fuel. Near each end of the element sufficient swelling had occurred to fully utilize the capabilities of the fluted shape. The cause of the high localized swelling near the ends of the element has not been determined.

N-Reactor Fuel Support Development. Fatigue curves were nominally completed for 1/4, 3/16, and 1/8-inch wide tube-tube inner supports. Endurance limits expressed as reciprocating load for the three sizes of supports are approximately 280, 170, and 70 pounds, respectively. Fatigue data are currently being developed for the target fuel support assembly.

Target Element Development. A study to determine the effects of a high temperature reactor excursion on Zr-2 clad Al-Li alloy target elements is continuing.

Examination has been completed on an irradiated Al-Li target element double canned in aluminum and Zr-2 that failed during an ex-reactor heating test. This test did not follow the pattern of previous runs in that failure was observed at a lower temperature and two areas of failure were observed. Metallographic examination has established that the core in the region of both failures had melted, and both ruptures had occurred where target support tabs had apparently been torn off before the heating test.

Two samples have been provided to evaluate the possible use of a high cross section poison in the center of a target rod that would be dispersed when a target element is overheated. Irradiated target rods of Al-Li alloy were drilled to provide 0.125 inch diameter axial holes which were then filled with boron and boron carbide and recanned in Zr-2. Reactivity measurements before and after ex-reactor heating to rupture will establish whether or not dispersion of the buried poison will compensate for the loss of Li-6. An additional test element using a 50% Cu - 50% Cd alloy as buried poison is being prepared.

Elevated Temperature Reactions Between Cadmium Alloys and Zirconium. Previous studies on elevated temperature reactions between various poisons and Li-Al target core materials have shown a possible reaction between the cadmium alloys and the zirconium cladding. In order to determine if any reaction exists, several zirconium cans containing pure cadmium and several containing a Cd - 50% Cu alloy were welded closed and will be heated to various times and temperatures. Half the cans were in the bright-etched condition and half were autoclaved to also determine the effect of the oxide film on the reaction.

Compaction of Lithium Aluminate Powder. Lithium aluminate powders have been compacted to 72% theoretical density by cold pressing and 83% theoretical

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density by hot pressing at 500 C and 800 C. It is planned to compact these materials to higher densities by pneumatic impaction at temperatures up to 1350 C.

2. Corrosion and Water Quality Studies

High Temperature Oxidation of Zircaloy-2. Interest has recently been expressed in the high temperature oxidation of Zircaloy-2 by water vapor. Zircaloy-2 coupons (1 x 0.5 x 0.03 inch) have been exposed to a 25 torr water vapor atmosphere at temperatures of 900, 1000, and 1100 C. The kinetics of reaction at each temperature have been expressed by a parabolic fit (i.e., the weight gain expression is $[\Delta W/A] = kt^{1/2}$). Assuming parabolic kinetics, the rate expression for reaction at these higher temperatures is

$$k \text{ (mg}^2\text{dm}^{-4}\text{min}^{-1}\text{)} = 1.14 \times 10^{12} \exp (-46,000/RT).$$

At each of these temperatures it was noticed that breakaway did not occur and that the scale formed was adherent. The thin samples did deform during testing; the 1100 C sample was brittle, while the 900 and 1000 C samples were ductile at the end of the exposure.

Corrosion Evaluation of Hydrided K-Reactor Zircaloy Process Tubes. It was found that the oxide thickness on K-Reactor Zircaloy-2 process tubes was not thick enough to be measured by metallographic techniques. Anodizing methods for Zircaloy-2 were then investigated as analytical tools for oxide thickness measurements.

In 1% NaOH solution a direct relationship exists between the anodizing voltage and oxide thickness on Zircaloy (0.049 mg/dm²/volt). Anodization of Zircaloy will not occur until a sufficient potential is applied to overcome the electrical barrier formed by an existing oxide film. Therefore, oxide thicknesses on Zircaloy could be measured by determining the potential required to overcome the electrical barrier formed by the existing oxide and converting this value to equivalent oxide thickness.

To date, results have been obtained by this method on Process Tube 3075. Two pieces of this tube have been measured--one from the heavily hydrided portion and one from the in-flux portion. The respective oxide layers were 0.25 mg/dm² and 0.5 mg/dm².

The oxide layer of 0.25 mg/dm² for the hydrided portion of the tube was measured on the outside diameter. It was not possible to anodize the hydride layer on the inside diameter.

N-Reactor Fuel Element Rupture Tests. The rupture rate and severity following a fuel element cladding defect is being evaluated in the Irradiated Rupture Prototype (IRP), a shielded high temperature loop.

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Special size KER-irradiated N-Reactor prototype fuel elements are pre-defected in the Radiometallurgy Facilities and placed in a shielded test section which can be attached to the loop. After the water temperature in the loop is at the approximate surface temperature of typical fuel elements in the reactor, the shielded test section is valved in and the water contacts the defected fuel element. This water temperature is maintained until the fuel element releases core material, as determined by activity monitors. The water temperature is then lowered at the N-Reactor shutdown rate, about 3 C per minute, following a 5-minute hold at 300 C. The amount of core material release is followed during the test by activity monitors and is determined after the test by weight measurements. Other pertinent information is obtained by visual inspection and measurements before and after the test and by continuous activity measurements in different portions of the loop.

Five tests have been performed (during this reporting period) with KSE-5 elements (single-tube, thick-walled, 1.6% enriched to evaluate swelling behavior at high uranium temperatures) with a single 0.025" hole in each. The tests were performed with fuel elements containing two different core additions to compare in-reactor swelling rates. These were the standard N-Reactor fuel composition with 120 to 140 ppm of both iron and silicon added to the uranium and the second type was with 400 ppm iron and 800 ppm aluminum added to the uranium core.

The first two IRP tests used elements with essentially the same exposure (~ 1300 Mwd/ton) and temperatures (470 C maximum fuel temp) but each with the different core material. Both of the elements failed in a similar manner. Each had an incubation period (time at temperature before activity release) of 145 to 168 minutes and preliminary weight losses of 4 grams before cleaning. The rupture consisted of a single raised and torn blister about 5/8 to 3/4" in diameter.

The next two elements were exposed to 1670 Mwd/ton and had a maximum fuel temperature of 580 C. The element with the iron and aluminum core additions had a 255-minute incubation period and a 5-gram weight loss. The rupture again consisted of a single raised and torn blister about 3/4" in diameter. The standard N-Reactor core addition element ruptured quite severely. After an 81-minute incubation period, the activity buildup in the loop filter was quite rapid and reached 65 R/hr at the end of the test. Preliminary uncleaned weight losses for the element was 133 grams. The rupture had extended for 6" in length and about 2/3 of the way around the circumference of the element for the 6" length. This ruptured area was covered with many raised and torn areas. Considerable difficulty was experienced in removing the fuel element from the shielded test section due to the swelling.

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The fifth fuel element exposed for 2650 Mwd/ton and a maximum fuel temperature of 566 C, and with iron and aluminum additions has not failed after a 200-minute incubation period. This test is continuing.

Reprecipitation and Deposition From Decontaminating Solutions. Two proprietary reagents were tested to determine the changes in appearance and amount of redeposition from decontaminating solutions held at operating temperature for extended periods of time. Samples of solutions containing coupons of carbon steel and stainless steel were heated to operating temperature for 66 hours.

A solution of a proprietary oxalic acid solution (Turco 4521), at a concentration of 60 g/l and operating temperature of 75 C, darkened with time. Precipitation was noticed first after 66 hours and increased in amount until the end of the test.

A proprietary sulfamic acid solution (Turco 4306-D), at a concentration of 90 g/l and a temperature of 61 C, also darkened with time. No precipitation was observed while the solution was heated, but upon cooling, a precipitate formed in the bottom of the sample bottle. The inhibitor had not given any indication of separation in the heated solution but was observed to be floating in a cooled sample drawn after 42 hours at temperature.

Evaluation of Film Removal from N Steam Generators. Two steam generators at N-Reactor had been decontaminated using a 2-step procedure alkaline permanganate, followed by Turco 4306-D, a proprietary sulfamic acid solution. Poor technique resulted in excessive dilution of the alkaline permanganate, and the films and contamination were only partially removed from the stainless steel tubes. A visual examination of several pieces of tubing removed from the steam generators revealed that partial cleaning and film removal had been obtained. The film was a reddish-brown color instead of the normal black color. In areas near the tube sheet, the film had been removed. In areas further from the tube sheet (between Baffles 2 and 3), the film was partially stripped and still further from the tube sheet (between Baffles 4 and 5), the film appeared to have only changed color from black to reddish-brown.

Fretting Tests on N-Reactor Fuel Elements. Two special N-Reactor fuel elements, assembled with inner (buggy) springs having spring constants 40-70% of that normally used were exposed in the N-Reactor test section of TF-7. The purpose of the test was to determine if relative movement between the spring and outer fuel element would result in fretting corrosion of the fuel element cladding. Temperature and flow rate during the 125-hour test were 277 C and 32 gpm, respectively. Amplitude (peak-to-peak) and frequency of vibration (resulting from externally applied vibration) at the approximate midpoint of the test section process tube were 0.001-0.002 inch and 54-68 cps, respectively. Visual examination after exposure revealed no fretting corrosion.

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Chloride Concentrations in Crevices in N-Reactor Steam Generators.

An extensive tube examination program is being conducted to monitor the severe intergranular attack previously found in the N-Reactor steam generators and to determine if new types of corrosion are occurring. As part of this program, analyses for chloride were performed on the OD of 20 tube ends removed from the tube sheets during the March 1965 outage and the data were compared with data obtained from previous analyses. Present chloride concentrations are 0.03 to 0.13 micrograms/cm² now, as compared with concentrations of 0.95 to 1.9 micrograms/cm² in 1963. There were no obvious differences in chloride levels among the six steam generators sampled or between leaking or nonleaking tubes. X-ray fluorescent measurements indicated the chlorides were uniformly distributed over the tube surfaces. The chlorides were distributed primarily on the outer surfaces in 1963, but are not occluded deep in the corrosion product films.

It is concluded that the chlorides that were originally present from the manufacturing process are being leached out and that chlorides from the water are not concentrating in the crevice areas. This is fortunate because if the chlorides had been concentrating in the crevice area, this could possibly lead to chloride stress-corrosion cracking. Whether or not the chlorides found were a factor in the intergranular attack in the crevice area is not known.

Hydride Formation in Zircaloy Tubes. Severe hydriding has occurred in a number of the Zircaloy process tubes in the K-Reactors and may require replacement of many tubes unless the causes of this hydride formation are identified and ways are found to repair existing damage and prevent further hydride formation. Factors which may contribute to this hydride formation include: (1) damage to thin Zircaloy oxide layer from scraping by discharging fuel, (2) deposition of aluminum on bare Zircaloy surface with subsequent corrosion reaction to produce hydrogen, (3) fretting damage of Zircaloy oxide from vibratory contact with aluminum spacers, and (4) slight possibility of fluoride decontaminant effects on the Zircaloy surface. Present investigations to determine the cause of this Zircaloy hydriding problem include fretting tests, exposure and analyses of pretreated coupons under typical reactor conditions, and corrosion probe development.

Fretting may be the major or a contributory cause of the attack. The spacers are relatively light in weight and under heavy load. External vibration of the tube, if present, may cause true fretting with destruction of the protective ZrO₂ film. This could then lead to accelerated attack of the bare metal by the coolant or even to galvanic attack. Fretting is being evaluated in 1706 KEL Laboratory in a special test facility designed to simulate the small movement under heavy load. The samples are mounted in a heated tank, maintained at reactor effluent temperatures, and relative movement between the aluminum spacers and the Zr-2 process is achieved by externally applied vibration. The

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test is scheduled to continue for 18 weeks or nine operating periods. The hydride concentration of sections of the Zr-2 process tube sample will be determined after each operating period.

Pretreated coupons are being exposed to pH 6.6 process water in out-of-reactor facility TF-20 at 120 C. First samples, discharged after 360 hours, are being examined visually and analyzed for hydride. Similar pretreated coupons were installed in KE-Reactor tubes for exposure to standard process water and to water at different concentrations of pH, dichromate, and peroxide. Zircaloy coupons were discharged from KE single-pass tube 2952 after 14 months of exposure in 130 C process water. These samples and other Zircaloy samples exposed 3 to 12 months in other single-pass facilities will be analyzed for hydriding.

CO₂ for pH Control. Testing of pH adjustment to 6.6 by CO₂ addition is being performed in the 1706 KE in-reactor twin-tube facilities to evaluate effluent radioactivity and corrosion effects. Laboratory data have shown reduction in aluminum and carbon steel corrosion under these conditions. Savings in chemical costs might be possible by use of stack gas CO₂ to replace sulfuric acid for pH adjustment of reactor cooling water. Coupon corrosion samples and corrosion probes were installed in 1706-KE facilities SP-3, 4, X & Y for exposure in CO₂ treated water. Exposure periods will be one and three months, respectively.

3. Gas-Atmosphere Studies

Burnout Monitoring. Since the early part of January 1965, F-Reactor has operated on Production Test IP-725, which permits a 50 C increase in graphite temperature above the 735 C limit. On the basis of acceptable rates from oxidation monitors discharged February 3 and March 30, 1965, approval was obtained to raise the temperature limit another 50 C. At the next reactor startup, probably in mid-April, this supplemental production test will become effective.

Profiles of the burnout rates showed the absence of any O₂. The only peak appeared near the stack centerline where the temperature and the flux were a maximum. The peaks for the two tests were ~1% per 1000 operating days (%/KOD) in Channel 1067 and ~2%/KOD and ~3%/KOD for the first and second tests, respectively, in Channel 3580. In both tests monitors which were protected from the reactor gas atmosphere by a 3/8-inch thick wall of graphite oxidized at the same rate as exposed monitors, indicating a homogeneous reaction.

Monitors in Channels 1889 and 1960 at C-Reactor from January 25, 1964 to March 7, 1965, had rates below 0.6%/KOD. There were no indications

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of the presence of O₂, and the protected and exposed monitors had equivalent burnout rates.

Graphite-Zirconium Compatibility Loop. Tests Nos. 10, 11, and 12 have been completed in the Graphite-Zirconium Compatibility Loop at a maximum temperature of 820 C.

Test No. 10 operated for 455 hours at a water-vapor pressure of 4500-5000 ppm. The buildup of gaseous products was followed by gas chromatography. Within 48 hours of startup the H₂ and the CO had exceeded 1%. A mass spectrometer analysis taken at that time showed 1.43% H₂, 1.01% CO, 0.26% CO₂, and O₂, N₂, and CH₄ all less than 0.01%. These results, together with a few analyses made during Test No. 9, suggest that the water-gas shift reaction:

is being catalyzed. Appearance of the bar, especially where it contacts the Hastelloy-X, indicates that the Hastelloy-X may be catalyzing this reaction.

In Test No. 11 the water concentration was less than 80 ppm and hydrogen 1000 ppm. This test was designed to see if hydriding would occur under conditions where small lab experiments predicted hydriding. Hydriding was observed and will be reported elsewhere. Graphite oxidation was negligible ($<10^{-7}$ g/g/hr) as expected.

In Test No. 12 the carbon dioxide concentration was approximately 2% and water less than 50 ppm. Oxidation rates of 5×10^{-7} to 5×10^{-6} hr⁻¹ were observed for the surface samples. These rates are comparable to those obtained in Test No. 10 for the high-water-vapor concentrations. The carbon dioxide oxidation was more uniform over the interior samples than the water-vapor oxidation.

Gas-Graphite Reactions. The thermal reactivity of N-Reactor graphite (TSX) with dry air at atmospheric pressure was determined in the temperature range from 625 to 727 C. The samples were oxidized from 10 to 50% weight loss. The rate can be expressed by the equation,

$$R \text{ (g/g/hr)} = (2.29 \pm 0.04) \times 10^{10} e^{\frac{-51.8 \pm 1.9}{RT}}$$

The standard deviations for the pre-exponential factor and the activation energy are shown in the equation. The data were collected from 11 different samples and at seven different temperatures for a total of 21 individual determinations. The value for the activation energy agrees well with the value of 49.9 kcal/mole previously reported for the temperature range 497 to 630 C.

A comparison of earlier work and these data suggests that the reaction between TSX graphite and O₂ in the temperature range from 497 to 727 C

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occurs uniformly throughout the whole sample, i.e., the reaction is in Zone I where the rate is independent of the surface-to-volume (S/V) ratio. The sample geometry in the earlier work was a parallelepiped with dimensions from 2 x 8 x 20 mm and a S/V ratio of 1.3. The geometry in this latest work was solid cylinders with dimensions of 0.43 dia. x 4" long and 0.43" x 1" long with S/V ratios of 9.9 and 11.4, respectively.

At 630 C the reaction order with respect to the O₂ concentration was previously found to be approximately 0.5 over the concentration range from 0.2 to 1%. Using the value for the reaction order and extrapolating the reaction rate to the concentration of O₂ in air, excellent agreement was obtained with the reaction rate measured in this latest work.

4. Process Tube Development

Irradiated Tube Testing Facility. A report entitled "Irradiated Burst Test Facility," BNWL-6, describing the facility and its testing capabilities, was issued this month. This test facility, which became operational in March 1964, has operated favorably since.

K-Reactor Welded Zircaloy-2 Process Tubing. A series of burst tests at room temperature and elevated temperature was made on sections of welded Zircaloy-2 tubing for the Hanford K-Reactors. In general, the strength of the tubing compares favorably with extruded and drawn tubing. At room temperature, failure is in the weld zone and at 300 C the greatest strain is in the welded heat affected zone, but failure is opposite the weld zone. Report BNWC-51 describing the tests has been issued.

5. Thermal Hydraulics Studies

N-Reactor Boiling Burnout Studies. Fourteen boiling burnout points were obtained in the study of heat transfer conditions of co-producer fuel elements for N-Reactor. The experiments were conducted in the heat transfer laboratory with an electrically heated model having the geometry of the co-producer inner annulus; i.e., a horizontal coolant annulus with a heated outer wall and an unheated inner wall. The annulus width was 0.2765 inch with an inside diameter of the heated outer wall of 1.775 inch and an outside diameter of the unheated inner rod of 1.222 inch. The purpose of the present experiments is to investigate possible effects of a heated surface versus an unheated opposing surface on burnout behavior. Future experiments, with models having dimensions more closely matching the co-producer fuel, will investigate burnout behavior in the entire co-producer fuel and the effects of dimensional changes.

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The fourteen burnout points were obtained at 1500 psig with mass velocities of 0.5, 1, 2, 3, 4, and 5 million lb/hr-sq ft and with inlet temperatures of 590, 560, and 530 F (310, 295, 275 C). Outlet conditions at burnout ranged from about 25 F (15 C) subcooling to 10% steam quality. Thirteen of the burnouts were shown by abnormal temperature rises indicated by thermocouples used as burnout detectors. These burnouts occurred at or near the downstream end of the 24-inch long heated section. The fourteenth point resulted in physical failure of the model at a point about 8 to 10 inches from the downstream end. The failure, a 1-1/4-inch long crack in a bulged, discolored area, occurred about two inches downstream of a set of ceramic supports separating the inner and outer tubes.

In general, the results of these experiments seem to agree quite well with those for a two-heated-surface model run previously. A preliminary examination of the data indicates that burnout heat fluxes were about the same for the two models, at comparable enthalpies and mass velocities. The enthalpy range covered in these experiments, however, was rather limited. Plans were made to repair the model and conduct experiments which will extend the comparisons to greater subcooling.

Hydraulic Tests. Visual flow tests were conducted in the Hydraulics Laboratory to determine the vibration response of an N-Reactor target element. The element is the innermost element of the N-Reactor fuel column and is supported in the uranium fuel piece by nine buggy-spring bumpers along the length and periphery of the target element. The initial design value for the spring constant was to be 0.002 inch deflection per pound force but, because of etching, the value was reduced one-half. With such a low spring constant it was considered possible that the assembly might reach resonant frequency at the flows which would be encountered in the target element channel.

Tests performed in the laboratory with the 1.360 inch OD element (with rails) supported in a 1.345 inch ID pyrex tube indicated that excessive element vibrations would not be encountered up to flow rates of 35,000 lbm/hr (in-reactor mass flow rate = 33,600 lbm/hr).

The relative frequency and amplitude response of tube and element were measured using a strobe-light, reed tachometer, and grid microscope. High speed movies to frame speeds of 8500 frames per second also were obtained. Visual observation with the above equipment indicates 3 to 4 mils amplitude and 2100 cycles/min of tube and element vibration at 15,000 lbm/hr and 6 to 8 mils amplitude and 3200-3500 cycles/min at 35,000 lbm/hr. The latter frequency represents the approximate pump frequency.

Relative deflection between tube and element was difficult to measure and was not visually detected due to its small amplitude and the variable frequency response of the system.

6. Graphite Studies

N-Reactor Irradiations. The two capsules currently in the GETR, H-5-5 and H-6-5, are operating satisfactorily. The faulty connections made in the terminal box during the installation of H-6-5 have been corrected, and all thermocouples in both capsules are functioning properly. All sample temperatures are in agreement with previous capsule history.

The report, HW-81250 A, "The H-4, H-5, H-6 Irradiation Experiments: Irradiation of N-Reactor Graphite--Interim Report No. 1," on the data obtained from the first nine capsules in the program, H-4-1 through H-6-3, has been issued. Results are presented for samples irradiated to a maximum neutron exposure of 4.8×10^{21} nvt, $E > 0.18$ Mev. The TSX samples all showed continued contraction with increased neutron exposure, and to the exposures reported, the TSX graphite exhibited nearly the same contraction behavior as the reference CSF graphite in both the transverse and parallel orientations.

The N-Reactor graphite tests in the GETR will be terminated by June 30, 1965. Investigations are under way to determine the feasibility of irradiating these samples in the C-Area Reactor at the Savannah River Plant during the high-flux operation there. The fast flux ($E > 0.18$ Mev) is of particular interest for the graphite irradiations, and possible methods for determination of this flux by both calculational and measurements techniques are being examined.

7. NRD Shielding Studies

Shield Cask Code. The shield-cask code is presently being debugged. The code has been on the computer with a sample problem (a shield cask of rectangular geometry) and has output in the correct format; however, problems still exist in the calculational routines. The sample problem has an existing hand calculated solution. When the code yields a satisfactory solution to the sample problem, the code will be further checked by solving a cylindrical shield cask design problem for which a hand solution also exists. During this period a special isotope (isotopes other than isotopes produced from direct fission yield and subsequent decay) library was compiled for use with the code. Also completed were the buildup factor and mass attenuation coefficient libraries.

B. WEAPONS - 03 PROGRAM

Research and development in the field of plutonium metallurgy continued in support of the Hanford 234-5 Building operations and weapons development programs of the University of California Lawrence Radiation Laboratory (Project Whitney). Details of these activities are reported separately via distribution lists appropriate to weapons development work.

C. REACTOR DEVELOPMENT - 04 PROGRAM

1. Plutonium Recycle Program

Fuels Development

PRTR Fuel Element Performance. Two discolored areas on the cladding surface of the defect rod (DI-20) from PRTR FE-5237 were identified as zirconium hydride by radiometallurgical examination. The element failed in January 1965, after reaching an exposure of 1×10^{20} fissions/cc (3420 Mwd/ton fuel). The fuel contained 40 ppm oil, but this rod was reported to have had three discolored spots on the cladding after autoclaving. It has not been determined if the hydriding originated on the internal or external cladding surface.

The first 2 wt% PuO_2 element, containing three rods of salt cycle fuel material, was discharged from the PRTR rupture loop and examined. One band on the downstream end was twisted and was replaced. The element is ready for recharging into the reactor.

A salt cycle fuel rod was successfully irradiated in the MTR and is presently awaiting postirradiation examination.

Fuel Element Refurbishing. Two PRTR irradiated fuel elements were repaired in the PRTR basin. One element had a broken wire wrap which was successfully repaired. The other element had a number of broken and missing bands, and this element was rebanded with three new bands.

Fuel Recycle Pilot Plant. Design and procurement of remotized fuel element refabrication equipment, for initial recycle operation, is estimated to be 20% complete. Purchase requisitions have been issued on all capital equipment to be procured for this purpose from FY-1965 equipment funds. Installation is scheduled to begin as soon as FRPP (324 Building) construction progress will allow.

Spheroidization Studies. Agglomeration techniques for UO_2 and UO_2 -plus-additives are being evaluated to produce feed materials for vibratory compaction of fuel elements. Five different starting materials and processes have been studied to date.

1. Low density UO_2 fines and nuggets were agglomerated into spheres by ball milling using large (2-15 mm diameter) UO_2 chunks for the mixing action.
2. High density UO_2 fines and nuggets were agglomerated by the technique used in (1) above.
3. Calcium oxide and micronized UO_2 were combined and ball milled using large UO_2 chunks for the mixing action. The calcium oxide appeared to accelerate the agglomeration action.

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4. Micronized molybdenum and micronized UO_2 were combined using technique (3) above; however, the resulting agglomerates were rough. Subsequent rolling treatment on vibrating emery cloth produced hard, smooth agglomerates.
5. (a) Larger (2-6 mesh) spheroidized particles have been produced by adding a binder to micronized UO_2 forming large granules which were then rolled by hand in trays to generate spheroids.
(b) The same treatment as (a) except the starting material was a -300 mesh arc fused UO_2 and after the granules were formed, the green chunks were vibrated in a tray of micronized UO_2 .

All of the above material requires a sintering treatment as the final densification step. Final densities ranging from 74 to 98% of theoretical density have been produced. Particle sizes varying from +2 to -325 mesh have been made by one or more of the above processes.

PRCF Ultrasonic Crack Testing. Eight samples of 20 PRCF fuels were ultrasonically tested for cracks in the welded portion of the fuel rod at the clamped end. One small crack was detected in one of the samples. The remaining fuel rods in that batch were tested and another small crack was detected. Due to tight time schedules, test standards could not be fabricated for the PRCF fuel geometry so the exact size of the cracks is questionable. It is felt, however, that the cracks are insignificant for PRCF use and none of the fuel was rejected.

Fuel Cycle Analysis. A test case for the cost of impacted, vibrationally-compacted mixed oxide fuel elements has been successfully programmed through the Fuel Element Fabrication Code. The numbers submitted for the building cost, overhead costs, etc., are highly tentative. When more accurate values for the reject rates and overhead charges are substituted, the calculated costs will be more realistic. For purposes of comparison, a case is being prepared for a pellet fuel element.

PRTR High Power Density Core Fuel Elements. The maximum burnup on a UO_2 -2 wt% PuO_2 HPD element in PRTR is 3030 Mwd/tony with an average of 1775 Mwd/tony for the seven HPD elements presently in the core. These burnups have been accumulated at maximum rod powers of 19 kw/ft which produce fuel temperatures as high as 2600 C at the mid-plane of the elements.

Postirradiation examination of the center rod and rods from the six-rod ring from HPD FE-6000 (maximum rod power 19 kw/ft) indicates that the power generation was appreciably lower in these inner rods than in the outer rods from the 12-rod ring. This was also indicated by the lower gas release from the inner rods. No central void was formed at the mid-plane of the inner rods from the element and only a small tear-drop void

formed in the bottom of the rod from the six-rod ring. These fuel structural effects were expected since calculations indicate that the rod power generation rate in the six-rod ring and the central rod is only 55 and 42%, respectively, of the rod power generation rate in the 12-rod ring.

Preliminary calculations indicate that a plenum will be necessary to accommodate the gases released during high temperature operation of HPD elements to prevent excessive internal pressures.

Fuel Element Rupture Test Facility. There are plans to irradiate a HPD element in the FERTF at a maximum rod power of 22.5 kw/ft (515,000 Btu/hr-ft²). This power will yield fuel temperatures above melting with as much as 30% of the cross-sectional area molten.

Corrosion and Water Quality Studies

Boron Recovery From Used Resin. An effective (but not necessarily optimum) regeneration technique was developed to recover H_3BO_3 from the anion resin that will be used to remove H_3BO_3 at PRTR. The procedure utilizes Na_2SO_4 or H_2SO_4 as the regenerant and provides quantitative recovery of H_3BO_3 in 10 bed-volumes of regenerant. Recovery of H_3BO_3 from the regenerant can be effected by sulfate precipitation. An alternate technique utilizing NH_4OH as regenerant will be studied to attempt simplification of recovery from the regenerant. A simple, effective regeneration-recovery process will definitely be required if B^{10} -enriched H_3BO_3 is used as the chemical shim additive at PRTR.

H_3BO_3 Ion Exchange Studies. A laboratory model of the PRTR moderator and H_3BO_3 removal ion exchange system was assembled and operated under conditions similar to those proposed for the Phase 1 tests. Two tests were performed to demonstrate the H_3BO_3 control capability. The system was then disassembled and the boron concentration profile through the ion exchangers was determined. Results demonstrated that the system design is entirely satisfactory for proposed Phase 1 operations.

Additional work is in progress to determine resin boron capacity values under dynamic conditions and to evaluate the factors controlling boron elution in the resin beds. Results from these studies will be used to define the performance of the H_3BO_3 removal exchangers and the performance of the borated IX-3 exchanger during Phase 2 operations.

H_3BO_3 Prototype. Installation of all prototype components in the PRTR was completed. Design Acceptance Tests (using H_2O) are currently in progress and should be completed by the end of April.

Localized Corrosion in Shim Wells. A pilot plant corrosion test is in progress in the EDU Shim Test Loop to determine the corrosiveness of 140 F, 50 ppm H_3BO_3 to a PRTR shim rod and shim well. The shim rod is in

the lower position so that it is in contact with the shim well walls. A flow of 3 gal/hr is maintained past the shim rod. Examination of the shim rod after two and four weeks of exposure revealed very little generalized corrosion; however, numerous white deposits resembling aluminum corrosion product were observed on the shim well walls after four weeks. Since these deposits resemble the deposits which covered pitted areas during previous testing, the shim well will be removed for destructive examination. Corrosion measurements on coupons of aluminum and stainless steel after two weeks of exposure showed negligible penetrations.

High Radiation Levels in PRTR Primary System. PRTR primary system radiation levels have been increasing. Shutdown radiation data show that the buildup rate accelerated in August or September 1964, after being essentially constant since the PRTR decontamination. This accelerated buildup rate has continued into the Spring of 1965. Work has been started on defining the variables affecting PRTR radioactivity buildup rates. The following summarizes program progress during the past month:

- (a) Crud Levels. A crud probe installation has been used to obtain a primary system sample for radiochemical studies. Preliminary results show the presence of Co-60, Fe-59, and Sb-124. Other species are present, but they have not yet been identified. Since a quantitative study of coolant borne crud levels is dependent upon knowledge of flow, a flow measuring device is to be installed on the crud probe equipment at PRTR in late April.
- (b) Radioactive Films. Two nozzle cap coupons are being discharged to provide data on the identity of radioactive species retained in PRTR primary system films. Supplementary data are being obtained from PRTR components, but these sources are inadequate for all but qualitative measurements. Sufficient specimens for a detailed study of adsorbed activity and loose crud levels will not be available until the By-Pass Flow Loop becomes operational. Completion and operation of this facility has been postponed until June 1965.
- (c) Components. Inspection of various PRTR components continues. Several fiberglass filters from various PRTR streams have become available during the past month. Five appeared to be predominantly loaded with ion exchange resin. One was coated with a heavy black deposit which has been identified as organic material.

PRTR Crevice Corrosion Tests. Out-of-reactor tests to study corrosion in heated crevices, with geometries similar to those existing on the heat transfer surface of PRTR fuel elements, continued during the month. Microscopic examination of sections from the heated Test Assembly No. 2

(described in the March report) verified that the crevice geometry resulting from the addition of "cluster" bands to the Zircaloy-2 clad heating units is conducive to accelerated, localized corrosion.

Test Assembly No. 2 had been exposed in lithiated water at pH 10 for 60 days with a surface heat flux of 550,000 Btu/hr-ft² surface. From a preliminary examination, it appeared that corrosion was even more extensive than that associated with an initial specimen exposed to the same heat flux for 70 days. Sectioning showed that this was not the case; maximum heating-unit surface penetration, where "cluster" bands had contacted the heated length of the initial and second specimens, was approximately 0.007- and 0.004-inch, respectively. Microscopic examination of sections from Test Assembly No. 2 also revealed heating unit penetration where wire wraps had contacted the heated length. In some locations the penetration associated with wire wrap contact areas, up to 0.002 inch deep, was completely filled with gray-white corrosion product deposit. Corrosion of the "cluster" band was limited to crevice areas where the band contacted the heated length of the heating unit.

PRTR Corrosion Monitoring. Continuous monitoring of corrosion in various PRTR systems was proposed at the time the reactor was being built. This monitoring was to detect corrosion changes and provide a continuous history of corrosion in the shield, reflector, and moderator system. Provisions were made during construction for later installation of probes. Accordingly, four probes were installed in 1961: bottom shield inlet and outlet, reflector, and moderator. Two of these probes apparently did not provide any initial readings. One probe provided an initial reading but is no longer operable and one probe of 6061 aluminum is still operable and indicates an average corrosion rate of 0.2 mil per year in the reflector system. Replacement of the other three probes is now scheduled for the present outage and additional probe installation is planned for the May outage.

PRTR Pressure Tubes

Zr-Nb Pressure Tubing. Brittle fracture tests at room temperature on the 30% CW tubing were started this month. Tests employing a 1-1/2-inch long slot were completed. In the first test with a 1-1/2-inch long slot milled to a depth of 80% of the original tube wall thickness, failure occurred at an internal pressure of 5460 psi, and there was no crack propagation. This specimen was subsequently resealed and repressurized. Failure occurred at an internal pressure of 7060 psi. Crack propagation was substantially less than for Zr-2 in a cold worked condition.

An irradiated specimen having cold worked microstructure was tested for crack propagation resistance. The specimen failed at a nominal stress of 26,000 psi at room temperature. The specimen's average exposure was

1.2×10^{21} nvt. Another similar specimen had failed at a nominal stress of 24,000 psi, but it had an average exposure of 6×10^{20} . The 100% increase in neutron exposure is indicated to have produced an 8% increase in crack propagation stress. This is the second instance of an indicated increase in crack propagation stress accompanied by an increase in fast neutron exposure. In both cases the stress increase was of the order of 8% and the exposure increase was about 100%.

Reactor Components Development

Fretting Corrosion Investigation. Additional in-reactor tests have been conducted to evaluate zirconium concentration in the primary coolant system. As in the first test, the data show the primary system zirconium content as the steam pressure was increased from 335 psig to 365 psig and maintained for an extended time period. Primary bulk and channel 1653 coolant samples were periodically drawn for chemical analysis. There appears to be a definite increase in zirconium concentration with time, averaging better than twice as high in Channel 1653.

Inlet jumper vibration data from Channels 1653 and 1354 were taken during the test and the applicable traces analyzed. These data indicate that the amplitude of the Channel 1653 inlet jumper increased approximately 10%. When the steam generator pressure was increased to 365 psig, Channel 1354 initially decreased approximately 12.5% after which the amplitude increased slightly to reflect an approximate 10% decrease based on the amplitude of vibration at the start of the test. These amplitude changes are approximately one-half as much as occurred previously under similar test conditions. Similarly, the increases in zirconium content are considerably less than noted previously. Comparison of these data with those taken during the prior primary coolant pressure (temperature) effect investigations indicate that the ratio of zirconium concentration at a pressure of 365 psig, to that at 335 psig, varies directly as the ratio of inlet jumper motion of channel 1653 at 365 psig to that at 335 psig.

Fuel Re-use Study. Construction of the liquid sodium loop for the fuel re-use experiments is complete. Two hundred pounds of sodium have been charged into the loop storage tank and shakedown runs of the loop are in progress. The over-all Liquid Metal Facility has some remaining work to be done on peripheral equipment before it is complete.

Those systems of the sodium loop that it is possible to operate without circulating sodium have performed without difficulty.

Gasket Study. A test program for improving the performance of the spiral-wound asbestos-filled gasket seals on the PRTR pressure tube assemblies has been formulated (BNWC-30), and testing is under way. Low chloride asbestos, stress relieved stainless steel winding, and substitution of Inconel for stainless steel are being investigated as means of minimizing or eliminating stress corrosion cracking; thermal and autoclave treatments are being investigated as a means of avoiding the apparent gasket "relaxation."

The gasket testing station duplicates the PRTR nozzle cap, tube-nozzle and inlet jumper-tube closures. Five simulated PRTR thermal cycles have been completed on two sets of Inconel wound, and one set of stainless steel wound, asbestos-filled gaskets. On the inlet closure, there was no leakage from any of the gaskets until the 5th cycle when one of the Inconel gaskets leaked 2 ml/hr; on the tube-nozzle closure, the Inconel gaskets had no leakage, but the stainless steel gasket consistently leaked 0.5-3 ml/hr; and on the cap closure, the Inconel leaked 2-7 ml/hr, while the stainless steel essentially did not leak.

The holding torque of the bolts for each fixture was determined after the third thermal cycle. The torque on the bolts at the nozzle cap and tube-nozzle closures were the same as made at installation. However, the torque on the bolts at the inlet-tube closure of all three test stations had decreased from the original 45 ft-lbs to values between 26-45 ft-lbs with an average of 33 ft-lbs.

Design Analysis

Plutonium Utilization. Further examination of ZODIAC-2 input data for the calculation of Yankee Core 1 burnup has disclosed several minor errors; these have been corrected. In addition, the saturable fission products Xe-135 and Sm-149 were included to obtain a better estimate of the neutron spectrum during burnup. However, the net result of the changes made in the calculation was not sufficient to bring the reactivity loss with burnup into agreement with measured values. A search for additional discrepancies has been resumed.

Thermal Hydraulic Studies

Tests were run in the Hydraulics Laboratory to determine reasons why graphite has been lost from a crud sampler used to analyze for particulate matter in the PRTR flow system. The sampler consists of a porous basket containing 10-15 grams of powdered spectroscopic graphite which filters a 0.15 to 2.0 gpm stream of the process water operating at 1050 psig and 520 F.

During several of the sampling periods at the reactor, the graphite has been lost, apparently having been washed into the primary system. The

nature of the graphite loss was not determined; i.e., whether the graphite was being washed around leaky sealing gaskets and into the stream, through the porous stainless steel basket bottom, or whether it was lost through voids in the weld around the periphery of the porous bottom was unknown. Tests performed in the 189-D laboratory indicate that a gasket failure would be evidenced by visible indications around the sample holder and could be readily detected.

Tests run over a two-hour period at a pressure differential of 90 psi (reactor conditions) between the upstream and downstream portion of the basket showed no apparent graphite loss (by weight) at 10 C coolant temperature. At a pressure differential of 500 psi over a one-hour period, 0.9 gram was washed through the basket. The latter pressure differential, of course, represents a much more severe condition than would occur during operation of the reactor sampling system.

It is possible that, during the initial valving process in which the upstream valve is opened, the free expansion of the 520 F water at 1050 psig into the basket holder (which is initially at atmospheric conditions) produces a high velocity steam jet that breaks up the fine particles of graphite. Also, the rapid expansion of the water in the previously saturated graphite particles may, due to rapid heat addition and steam formation, contribute to the disintegration of the spectroscopic graphite.

Another possible reason for the loss of graphite could be a mistake in valving at the reactor since the wrong sequence could wash the graphite out of the basket.

2. Plutonium Ceramic Research

PuO₂-Stainless Steel Cermet Irradiations. Postirradiation examinations are being made of high and low density stainless steel-20 vol% PuO₂ cermets. The cermets were irradiated in the ETR at a calculated specific power of 3.86 kw/inch to 1.6% burnup of the fissile atoms. Only a few preliminary results of these examinations are available.

The high density (97.3% TD) cermet pellets operated with a calculated centerline temperature of 1300 C. No evidence of reaction between the PuO₂ and the stainless steel could be detected in the polished specimens. There are indications that some sintering occurred in the central portion of the pellets. In general, the high density cermet appears to be sound. A total of 6.07 cc's of gas with an activity of 50 mrad/hr was obtained from the capsule containing the high density pellets.

The low density cermet pellets operated with a partially molten core during irradiation. Calculated centerline temperature for these pellets was 1415 C. The four pellets in the capsule fused together. The structural changes which occurred in the low density cermets are analogous to those in irradiated UO_2 . A central void was formed which extended through three of the four pellets in the capsule. The region surrounding the central void contained a very dense matrix, free of voids and somewhat depleted in PuO_2 . The peripheral region of the pellet was only slightly changed from the as-fabricated condition but did contain evidence of sintering. There are no indications of gross dimensional changes. A total of 5.18 cc's of gas with an activity of 500 mrad/hr was obtained from the capsule containing low density pellets.

Measurements on cermet density and off-gas compaction are under way. A more detailed metallographic examination of the irradiated specimens is also being made--with particular attention to changes in the PuO_2 and reactions between the PuO_2 and the stainless steel matrix.

Production of High Purity $\beta\text{-Pu}_2\text{O}_3$. Six hundred grams of $\beta\text{-Pu}_2\text{O}_3$ were prepared by carbon reduction of PuO_2 . Sintered pellets of the reduced material were arc melted in a purified argon atmosphere to remove volatile impurities and to improve the density of the material. The density of a representative sample, after melting, was 11.36 gm/cm³ (99.2% TD). The arc melted buttons contained blue and greyish-yellow tarnish films which were determined to be PuO_2 with a lattice parameter of 4.923 ± 0.007 A from x-ray diffraction patterns of the button surfaces. Several buttons with metallic black surface films contained some $\alpha\text{-Pu}_2\text{O}_3$, but no PuO was found on the surface of these samples.

The prepared $\beta\text{-Pu}_2\text{O}_3$ is being analyzed for gas content, semi-quantitative impurity content, oxygen-plutonium ratio and percent plutonium content.

Plutonium Nitride. Synthesis of a stock of PuN for use in fabrication, property measurements, and irradiation experiments is continuing. Several procedures have been tested and an optimum procedure was chosen for use with presently available equipment. X-ray diffraction indicates oxygen contents running generally less than 0.4%. Lattice parameters show good consistency between batches. An apparatus to provide analysis for nitrogen by a modified Kjeldahl method is being developed.

PuN-SS Irradiation. $\text{PuN-20 wt\% SS}$ (type 316) pellets were pressed to 70% TD in preparation for a PuN-SS irradiation compatibility study. These pellets were left overnight in an oxygen contaminated nitrogen hood atmosphere during which time the PuN-SS pellets oxidized yielding

approximately an 8% growth in length and 2-4% growth in diameter. The dimensional changes were sufficiently large to preclude their placement in the stainless steel can intended as the primary container for the irradiation study. This rate of oxidation indicates that extraordinary care must be taken and some hood modification made to assure meaningful studies of plutonium nitrides or uranium-plutonium nitrides.

Plutonium Ceramics Metallography Facilities. Techniques for the metallographic preparation of PuN and nitride-containing specimens in general are being developed. The hood facility is operating on nitrogen and contains less than 1/2% free oxygen in two sections of the hood. The section nearest the air lock contains one to two percent O₂.

High Temperature Dilatometer. A plastic water line inside the vacuum enclosure ruptured causing burnout of the tantalum heating element. The system has been completely cleaned, and a heating element of a new design was installed. The unit is now back in operation with water filters installed to prevent plugging and overpressure of the plastic tubes. The glove box facilities for the dilatometer are about 75% complete.

It has been found that the ThO₂ sample holders being used are dimensionally unstable at elevated temperatures (about 1300 C) and are therefore unsuitable for high temperature thermal expansion work. An attempt to stabilize the ThO₂ with a high temperature (1750 C) long time (65-hr) soak in a hydrogen furnace resulted in cracking of the tubes. It is felt that the cracking did not result from thermal shock.

Phase Studies. Oxygen dissociation pressures in the Am-O system have been measured at 1014 C. The oxygen pressure in the system was fixed by externally controlling the decomposition of a cupric-cuprous oxide mixture. Equilibrium over the reduced americium oxides was approached from both low and high temperatures and occurred rapidly for O/Am ratios greater than 1.95. The dissociation pressures varied linearly between O/Am 1.985 and 1.966 and extended from 14.3 to 0.96 mm O₂ at these compositions, respectively. In the vicinity of AmO_{1.90}, approximately seven hours were required to attain equilibrium with an associated oxygen pressure of 6×10^{-3} mm Hg.

Some doubt exists as to phase relations in the PuO system below PuO_{1.75}. A recent British diagram, obtained by high temperature x-ray diffraction, is in disagreement with the earlier Hanford diagram in that a eutectoid is not shown at PuO_{1.70}. This construction is inconsistent with thermal expansion and electrical resistivity

results obtained at Hanford, Harwell, and Mol. In our earlier diagram, phase fields below $\text{PuO}_{1.70}$ were not rigorously investigated due to sample oxidation. Currently, DTA and high temperature diffraction techniques using extremely dry atmospheres are being developed to examine this portion of the diagram. Samples with O/Pu ratios below 1.8 have been prepared by reacting PuO_2 and $\text{PuH}_{2.7}$, both prepared from high purity alpha plutonium metal, at 1650 C in vacuum. A sample of composition $\text{PuO}_{1.555}$ showed three phases: $\alpha + \beta \text{Pu}_2\text{O}_3$ plus a FCC phase at room temperature. Additional compositions are presently under examination.

The Plutonium-Boron System. Nine arc-cast samples of Pu-B compositions between 50 and 96 at.% B have been annealed at 1600 C and slow cooled over a period of approximately 16 hours. X-ray diffraction analysis of these samples is in progress. Compatibility tests were conducted prior to annealing. Boron nitride displayed satisfactory compatibility with Pu-B compositions, but reactions of varying severity were observed with the following refractory metals and oxides: W, Ta, Mo, Al_2O_3 , ThO_2 .

3. Ceramic (Uranium) Fuel Research

Heating Characteristics of Pneumatic Impaction Cans. The first attempt to establish the heating and cooling characteristics of pneumatic impaction cans use in the fabrication of cermets was partially successful. The cans consisted of double walls of molybdenum separated by MgO insulators. Four W-5% Re - W-26% Re thermocouples were positioned half-way up the can: (1) between the outer molybdenum can and the MgO liner, (2) between the MgO liner and the inner molybdenum can, (3) 0.25-inch from the inner molybdenum can, and (4) in the center of the can. The inner molybdenum can was filled with 0.25-inch thick molybdenum discs.

The thermocoupled can was placed in the induction heating coil and heated while the thermocouples recorded the temperatures. An optical pyrometer was used to measure the outer surface temperature of the can.

Only two of the four thermocouples, Nos. (1) and (4), operated satisfactorily. The results showed that the temperature in the center of the can lagged the outer surface temperature by about 500 seconds (6 minutes). The temperature lag between the outer surface temperature and thermocouple No. (1) [on the inner surface of the outer molybdenum can] was only about 150 seconds (2.5 minutes).

When the outer surface temperature reached about 1400 C, a longitudinal break occurred in the can, and it was necessary to discontinue heating. Coincident with this break, thermocouple No. (1) ceased to function, probably because of oxidation. The can was left in the induction coil

for about five minutes during which the central temperature dropped only 50 C and was then transferred in 15 seconds to the cold impaction die. While in the die, the central can temperature (thermocouple No. (4)) dropped at the rate of about 2 C per second. The results of this experiment indicate that:

1. Cans should be heated for at least 15 minutes after the external surface reaches the desired temperature. A heating time of 30 minutes is recommended.
2. Total heater-die transfer time plus die residence time prior to impaction should be recorded for each shot. Central can temperatures should be corrected by the observed temperature loss-rate of 2 C/second.

Relationships which were not determined because of the break in the can, and which are sufficiently important to justify a second attempt, include:

1. Relative temperatures on either side of the MgO insulators.
2. Relative temperature loss-rates near outer can surface compared to the center of the can when the can is in the die.
3. Actual time to reach temperature equilibrium throughout the can during heating.
4. Behavior at higher temperatures (to 1800 C).

Knowledge of these factors will permit accurate estimates of the impaction temperatures and of the can temperature uniformity.

Pneumatic Impaction of Pure Tungsten. In a study of the basic mechanism of pneumatic impaction, plates of pure tungsten, both 5 μ powder and -150 mesh spheres, were prepared by pneumatic impaction at 1400 C. The plates showed some laminations and cracks but will be satisfactory for obtaining density, metallography, and bend strength specimens. Plates of the same materials will be produced by pneumatic impaction at 1600 C and 1800 C. The aim of this work is to establish the optimum temperature for bonding tungsten by pneumatic impaction. Subsequent experiments will be concerned with varying the impaction pressures and using post-impaction heat treatments.

Pneumatic Impaction of Beryllium Powder. Fine (-17 micron) beryllium powder has been obtained. It will be loaded into a pneumatic impaction can and impacted at about 1000 C. A cylinder about 2 inches in diameter by 2 inches high will be made so that both vertical and horizontal tensile specimens can be machined. It is hoped that pneumatic impaction

can be used to produce beryllium bodies having a random small grain structure which will exhibit tri-axial ductility. Most commercial methods currently being used produce preferred orientation of the grains and uni-axial or bi-axial ductility characteristics.

Pneumatic Impaction of BeO and BeO-containing Fuels. The pneumatic impaction process is currently being investigated as a means for densifying BeO, BeO-UO₂, and BeO-PuO₂. Initial effort in this area is devoted to the pneumatic impaction of BeO and BeO-UO₂, to be followed by an evaluation of the latter's fission product retention capabilities. Other property measurements, such as strength and internal friction, will be determined for both materials where appropriate and necessary to evaluate fabrication characteristics. As information is acquired, emphasis will be shifted to fabrication studies of BeO-PuO₂ fuels for reactor concepts such as the EBOR (Experimental Beryllium Oxide Reactor).

Two pounds of Japanese NGK-type, CF-grade BeO were supplied by General Atomic for pneumatic impaction studies. This material reportedly sinters to high density at low temperatures (1450 C). As a result, the grain structure remains small, so that relative strength, structural integrity, and fission product retention may be enhanced. However, the initial submicron size BeO particles agglomerate easily, and tend to sinter inhomogeneously, so that there are regions of extremely high density (almost transparent) and regions of lower density which permit the fission gases to escape. The pneumatic impaction process may alleviate the agglomeration problem, while yielding dense bodies of small grain size.

In an initial test the BeO powder (as-received) was pressed into the cavity (0.75" OD by 1.5" long) of a mild steel die, and the assembly inserted into a 2" OD stainless steel impaction container. In turn, this container was placed inside a 2.5" OD stainless steel can, the annular area containing a MgO thermal insulation sleeve. The assembly was then evacuated, heated to 1200 C, and impacted at ~300,000 psi. Post-examination of the impacted BeO pellet revealed extensive laminations throughout the pellet length. A dark gray (~0.1-inch thick) zone, circumferentially surrounding the BeO, presumably was a reaction product of the BeO and mild steel die. A surface of the pellet was polished and etched for periods up to 1.5 hours in a cold, 50% HF solution, but microstructural features were difficult to distinguish. The latter may be due to the presence of some transparency, since the BeO pellet appeared extremely dense.

The laminar cracking of the above BeO pellet is thought to be a result of differences in the thermal expansion of BeO and mild steel and/or springback of the more ductile mild steel after impaction. To alleviate this possibility, current effort is being made to impact BeO pellets in an aluminum- or magnesium-oxide matrix. These oxides have more desirable thermal expansion characteristics and are expected to act as a buffer during springback.

Sodium Bonding Facility. Equipment is being designed and fabricated for use in an experimental sodium bonding facility. This includes a sodium extruder, sodium outgas furnace, and first stage bonding furnace under construction with a final stage bonding furnace under design. A preliminary equipment layout in the existing hood designated for the sodium loading facility has been completed. Detailed drawings will be made to incorporate the necessary hood changes.

UO₂-PuO₂ Fast Fuel Development. Approximately 8 Kg of UO₂-20 wt% PuO₂ fast reactor fuel is being prepared for irradiation evaluation in EBR-II. The high density mixed oxide is being prepared by ball-milling the powders to obtain a homogeneous mixture of small particles followed by pneumatic impaction to obtain a high density feed material suitable for vibrational compaction. High temperature vacuum outgassing (900 C for 4 hours at 10⁻⁴ mm of Hg) of the ball-milled powders effectively removes adsorbed gases which cause low impaction densities with the high surface area powders. The vacuum outgassing treatment also reduces stoichiometry to avoid excess oxygen in the fuel material. Analytical results on the first 1 Kg batch of UO₂-20 wt% PuO₂ processed by this technique were O/U ratio 2.007, gas content 0.036 cc/gm, impacted density 98-99% TD.

Effects of Atmosphere on the Electrical Resistivity of UO₂. A single crystal UO₂ specimen (1/4-inch diameter x 1-1/4-inch long) which had been heated in argon containing a slight oxidizing impurity was reheated to a constant temperature of 860 C in a reducing atmosphere (argon - 8% hydrogen). The electrical resistivity was observed to increase with time from an initial 1.6 ohm-cm to a saturated value of 8.0 ohm-cm in approximately 8 hours. A plot of log resistivity versus time showed a linear relationship.

Reproducible resistivity versus temperature data can be obtained to 1600 C when the measurements are made in the reducing atmosphere.

UN-20 wt% PuN Irradiation. The third and last irradiation capsule containing UN-20 wt% PuN pellets (GEH-14-618) has arrived at the Radiometallurgy Laboratory for postirradiation examination. The calculated burnup on the fuel in this capsule is 12.8 x 10²⁰ fissions/cm³.

X-ray diffraction analysis of the affected zone in the 304L stainless steel cladding used in a previous capsule (GEH-14-616) has shown no detectable amounts of any foreign phases. Further methods of analyses are being carried out.

Electron Microscopy of Irradiated UO₂. Specimens of sintered UO₂ used for long-term low temperature irradiation studies have previously been examined by electron fractography-replica methods and changes induced

by the irradiation correlated with physical property changes. To provide further microstructural information, polished surface electron microscopy of the same specimens is in progress.

Radiation levels from the mounted specimens are now low enough to permit processing them in the Ceramics Research Laboratory, where an excellent polish was achieved. Electron microscope replicas are in preparation. Microhardness determinations are being repeated on the repolished specimens.

Metallic Impurities in Mixed Oxide Fuel. Representative samples of typical lots of mixed oxides, used as fuel for PRTR, EBWR, and Saxton rods, were analyzed for metallic impurities such as Pb, Sn, and Zn. In general, impurity levels were about the same in all cases.

Some PRTR samples were representative of fuel elements now in the reactor that have accumulated exposures greater than 3000 Mwd/tony. Impurity levels (ppm) were: Sn 20-30, Pb 1-5, and Zn less than 50, which is the reportable lower limit of the emission spectrographic method.

Materials and Information Exchange. Twenty single crystal specimens of UO_2 were prepared, characterized, and shipped to Japan for the Central Research Laboratory, Toshiba, and the Research Laboratory, Atomic Fuel Corporation, for research studies and information exchange.

Requests for UO_2 crystals were received from the Watson Research Center, IBM Corporation, and the National Magnet Laboratory at M.I.T.

4. Basic Swelling Studies

Irradiation Program. A controlled pressure-temperature capsule containing six high purity uranium, two U + Fe-Al, and two U + Fe-Si specimens has continued to operate satisfactorily in-reactor at 1000 psi and 625 C. During reactor shutdown, this capsule is controlled at 1000 psi and 400 C.

Two controlled pressure-temperature capsules which will operate in-reactor at 500 psi and 450 C and 575 C, respectively, were assembled. One was shipped to the reactor for charging, and the other is being stored for future irradiation.

Postirradiation Examination. The density measurements made on uranium specimens removed from a swelling capsule irradiated at 575 C and 1000 psi to 0.15 at.% B.U. reveal that this pressure reduced the amount of swelling in high purity uranium by factors of 5 to 15 over that typically observed in uranium irradiated at low pressure. U + Fe-Al and U + Fe-Si alloy specimens exhibited reductions by factors

of 2 to 5. The effectiveness of this 1000 psi external, hydrostatic pressure is significantly less at 575 C than was the case for 450 C. At 450 C the reduction for high purity uranium was by factors of 50 to 100.

Preliminary metallographic examination corroborates the density measurements. The as-extruded high purity uranium specimen shows extensive grain boundary porosity (grain boundary tears were present in one end of the sample) and a few large pores scattered throughout the matrix. None of the crystallographically aligned microtearing typical of low pressure irradiation at 575 C was present. The 1000 psi pressure is effective in suppressing the aligned tearing at 575 C but not the grain boundary porosity and tearing. This behavior substantiates the concept that defect migration and agglomeration processes are responsible for swelling in this temperature range.

5. Irradiation Damage to Reactor Metals

Alloy Selection

Several nickel-base alloys are being studied to determine the effects of irradiation upon their mechanical properties. Specimens of Inconel 600, Inconel 625, Inconel 702, Inconel 718, Incoloy 800, Hastelloy N, and Hastelloy X-280 have been irradiated at 50, 280, 580, and 740 C and are currently being tested in tension in the temperature range of room temperature to 650 C.

Specimens of Hastelloy N irradiated at 280 C to an exposure of 1.03×10^{20} nvt ($E > 1$ Mev) and tested at room temperature and 650 C show a marked effect of test temperature. For the case of the room temperature tests, a 70% increase (to 74,750 psi) in yield strength and a corresponding 27% decrease (to 49.5%) in uniform elongation was found. The corresponding values at the 650 C test temperature, 44,600 psi and 6.1%, respectively, show that a marked decrease in yield strength and uniform elongation occurs at the higher test temperature. Similar results have been found for other solution hardenable nickel-base alloys.

Specimens of Inconel 625 have been irradiated at 280 C and tested at 650 C. These specimens were irradiated at exposures to 1.46×10^{21} nvt ($E > 1$ Mev) in the solution treated and aged condition. Results of the tests indicate little change in strength at exposures to 5.68×10^{20} nvt. At an exposure of 1.46×10^{21} , the data indicate a reduction in both strength and uniform elongation as compared to the lower exposure data.

Specimens of Inconel 718 irradiated at exposures of 1.03×10^{20} , 1.57×10^{20} , and 3.2×10^{20} nvt ($E > 1$ Mev) at 280 C were tested at 650 C. Data from these tests show an increase in yield strength from 87,440 psi at the lowest exposure to 138,610 psi at the highest exposure. The corresponding percent uniform elongation values were 5.21% and 0.85%, indicating a decrease in ductility. Similar effects were found for specimens irradiated at 50 C to exposures of 1.8×10^{20} and 1.0×10^{21} nvt and tested at 650 C.

To determine the effects of irradiation in a liquid metal environment, a temperature controlled capsule has been designed for insertion in the ETR. A bench model of this capsule has been fabricated and a safety circuit and temperature control system installed. Final assembly of the equipment is now being completed with actual operation of the capsule scheduled late this year.

In-Reactor Measurements of Mechanical Properties

Three in-reactor creep tests are currently in progress. Tests on annealed 304 stainless steel are being conducted at 750 C, 10,000 psi stress and 525 C, 30,000 psi stress. The 650 C test has accumulated 1200 hours of test time and exhibits a rate of 4.4×10^{-6} /hr, though this rate has not yet been established as a minimum. The 525 C in-reactor creep test and its control are in the primary stage of creep and minimum creep rates are yet to be determined. The third test, sponsored jointly by the AEC and the AECL, is being conducted on heat treated Zr-2.5 wt% Nb. Heat treatment consisted of heating specimens 15 minutes at 870 C, water quenching, and aging at 500 C for 24 hours, followed by air cooling. The in-reactor test was just recently started and a second stage rate cannot be established.

Another in-reactor creep capsule was recovered and sent to Radio-metallurgy for specimen removal. The creep test was terminated after specimen failure at test conditions of 650 C, 20,000 psi. This will be the first of the in-reactor creep specimens recovered at which test conditions were unchanged for the test duration.

Irradiation Effects in Structural Materials

Continuing efforts have been made to measure the crack velocity in a specimen undergoing catastrophic failure. These have been preliminary efforts with the major goal being the determination of the ability of a material to arrest a moving crack. To date the utilization of the Doppler effect to ascertain the crack velocity has proven somewhat unsuccessful due to the insensitivity of high-speed recording equipment. However, a new approach based on eddy current techniques is being studied, and equipment is presently being assembled for a trial run.

Irradiation Effects in Nickel-Base Alloys

The purpose of this program is to determine the effects of modified microstructures on the reaction to subsequent irradiation of nickel-base alloys. These structural modifications are made by pre-irradiation thermal and/or mechanical treatments and are screened by tensile and stress-rupture tests and metallographic examination on both control and irradiated material.

Twenty-four Hastelloy X-280 specimens irradiated at 280 C to a goal exposure of approximately 2×10^{20} nvt were discharged at the conclusion of ETR Cycle 70. These specimens, representing a variety of preirradiation treatments, will be returned to BNW for tensile and creep-rupture testing. Three quadrants (18 specimens) containing Hastelloy X-280 and Inconel X-750 will be irradiated during ETR Cycle 71. These specimens have been heat treated to give a range of microstructures. Metallographic samples of Inconel X-750, representing five different heat treatments, have revealed that the desired range of precipitate aging (from underage to average) was obtained.

Components for three creep-rupture units are being fabricated from nickel alloy material for testing in air to 925 C. Studies are being made to incorporate a completely automatic, hydraulic loading system as a further refinement of the prototype creep-rupture unit.

Damage Mechanisms

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

Capsules containing Ferrovac E and Battelle zone-refined iron have been sent to the reactor for irradiation to goal exposures of 5×10^{19} nvt and 1×10^{20} nvt. Iron specimens irradiated to 5×10^{18} nvt and annealed at 350 C for three hours have been sectioned, thinned, and examined in a JEM microscope. Distortion of the image by the magnetic field of the specimens is less severe than in the Philips microscope and should give better resolution. No spots were observed, however, in agreement with observations performed on the Philips.

A specimen holder for measuring resistivity ratios of single crystals has been designed and built. Troubleshooting of the high sensitivity resistivity apparatus is in progress and should be completed in the near future. A specimen holder for wire hairpin samples has also been designed and built and will be used to measure cold work recovery detectable by resistivity changes.

ATR Gas Loop Studies

Foil Insulation. The thermal conductivity tests on the foil insulation were completed, and the data are being analyzed. Preliminary checks showed that when a low temperature test point was run after the loop had been at high temperature the apparent conductivity of the foil was much higher than would be expected. Several test points were run which verified this indication. Gaps apparently formed at the joints in the insulation upon cooling down from a higher temperature, unless the test section was vibrated. The vibration would allow the insulation to settle into place and reduced the leakage of gas at the joint. A summary of results will appear in DCK-I-79.

Pipe Insulation. A topical report on thermal properties of Hastelloy-X felted metal insulation was prepared and is now ready for publication.

Model Loop Operation. During the month a bearing failure occurred in the eddy-current-variable speed drive in the frequency changer assembly. The bearing apparently had been run with some of the cooling water in the housing. The water washed the lubricant out of the bearing and caused the failure. One man-week was required to repair the unit.

Bypass Purification System. The bypass purification system on the model gas loop was improved by enlarging the low temperature charcoal and molecular sieve adsorber beds. This is the first step in making the system a complete cleanup system. A room temperature molecular sieve bed will be added when the parts arrive.

Helium Purification System. The high pressure helium purification system was placed in service following completion of repairs to the compressor. Helium was recovered from the 2500 cubic foot plastic balloon with the following results:

ppm	H ₂	O ₂	N ₂	CH ₄	CO	H ₂ O
Purity from Balloon	*	640	3000	33	*	8
Purity after 1st cleanup	3.5	1.3	1.9	*	*	7.5
Purity after 2nd cleanup	*	*	*	*	*	6.0

*No detectable amount from chromatograph, sensitivity is about 0.1 ppm.

Heater Design and Piping Modifications. All major components and material for fabrication of the concentric two-stage helium gas heater for the model loop have been ordered. Bids on the revised ceramic items have been returned with a saving of 80% of the original bids on these items.

A computer program was written to determine optimum insulation thickness, piping sizes and cooling water flow for the piping from the regenerative heat exchanger to the new heater and from the heater to the test section. The program also investigated the effects of cooling water flowing concurrent or counter current with the gas flow.

Dissimilar Metal Welds. The construction of the ATR gas loop will require the welding of thick wall sections of Hastelloy X-280 to Haynes 25 and AISI 316 stainless steel. The results were previously reported (1) on the welding procedure and properties of these weld combinations utilizing Hastelloy-W filler metal. Hastelloy-W has a long successful history in joining dissimilar high temperature alloys, but the results of short time elevated temperature tensile tests of wrought Hastelloy-W indicate a sharp loss in ductility near 1200 F (650 C). The results of tensile tests on wrought Hastelloy-X indicate a similar but less severe loss in ductility at 1200 F (650 C), and Hastelloy-X filler has also been successfully used in joining dissimilar metals. The effect of aging on the ductility of Hastelloy-X and Hastelloy-W weld metals aged at temperatures in the low ductility temperature range of 1200 F to 1600 F (650 to 820 C) has not been previously investigated. The aging is being done in inert gas at 1000 F (540 C) and 1200 F (650 C) for the AISI 316 stainless steel joints and at 1400 F (760 C) and 1600 F (850 C) for the Haynes 25 joints.

The relative ductilities of the welded joints are being determined from bend tests of 0.625-inch thick longitudinal weld specimens. The ductility is measured as the maximum elongation in the outer fibers of the material where cracking or failure was initiated. The bend testing of unaged, 500-hr aged, and 1000-hr aged specimens is complete. The tensile properties of aged Haynes 25 and Hastelloy X-280 base metals, and Hastelloy-X and Hastelloy-W weld metals are also being determined. The tensile testing of the 500-hr aged material is complete and specimens of 1000-hr aged material are being machined. Weld specimens for aging periods greater than 1000 hours are currently being aged. All of the unaged, 500-hr aged, and 1000-hr aged bend specimens initially failed in the weld metal.

(1) R. L. Knecht, Dissimilar Metal Welds of Certain Superalloys and Stainless Steels, HW-78575, August 1963.

As expected with aging, a loss in ductility and increase in yield strength was observed. The aging is considerably more sluggish in Hastelloy-W than in Hastelloy-X. Although both weld metals with a 500-hr age reach similar minimum room temperature ductilities, the tensile tests at the 1000 F (540 C) and 1200 F (650 C) aging temperatures indicate that Hastelloy-W has a more useful amount of ductility remaining than Hastelloy-X.

The effect of a 500-hr age at 1000 F (540 C) on the yield stress and ductility of Hastelloy X-280 is quite small and in good agreement with the results of a 100-hr age at 1000 F (540 C) reported by Levy and Wheeler.⁽¹⁾ The effects of a 500-hr age at temperatures greater than 1000 F (540 C) on yield stress and ductility of Hastelloy X-280 are relatively small when tested at the aging temperature and quite pronounced when tested at room temperature.

The above results of the tests on the weld metals and base metals aged 500 hours suggest that Hastelloy-W has better aging characteristics than Hastelloy-X when used as filler wire for joining Hastelloy X-280 to AISI 316 stainless steel and Haynes 25. This suggestion is based on considerations of the weld metal yield stress being larger than the base metals yield stresses, the relative sluggishness of weld metal aging, and the amount of useful ductility remaining in the weld metal and the base metals after an aging treatment. The tensile testing of the weld and base metals aged 1000 hours and greater than 1000 hours is continuing.

(1) Quarterly Progress Report - Metallurgy Research Operation - January, February, March, 1964, edited by J. J. Cadwell, HW-86269, April 15, 1964.

6. Nuclear Graphite Studies

High Temperature Graphite Irradiations. The second capsule, GEH-13-11, in the series of high temperature irradiations of graphite has completed two cycles of irradiation. Just prior to the current shutdown, two of the 12 thermocouples failed. Analysis of the cause of failure is presently under way.

EGCR Graphite Irradiation. The 10th capsule, H-3-10, in the series of long-term irradiations of EGCR graphite successfully completed the first cycle of irradiation in the GETR. All thermocouples functioned properly and sample temperatures agreed with those of previous capsules.

Effects of Additives on Graphite Contraction. Samples from the Contract DDR-136 were discharged after an initial exposure of 1625 Mwd/At_K at a temperature of 650 ± 50 C.

The samples were prepared in three sets: (1) Needle coke, pitch binder with the following additives: 5% Al, 5% Al₂O₃, 5% Al₄C₃, 4.5% Fe₂O₃-0.5% SiO₂, 4.5% Fe₂O₃-0.5% Al₂O₃, 4.5% Fe₂O₃-0.5% Co₂O₃. (2) Fluid coke, pitch binder with the following additives: 5% Ni₂O₃, 5% Fe₂O₃. This set also includes two purification schedules in which the purifying gas was introduced at either 2300 or 2900 C. (3) Needle coke, phenolformaldehyde resin binder with the following additives: 5% Fe₂O₃ and 5% Ni₂O₃.

In Set (1) at this low initial exposure, the sample with the Al additive contracted the greatest; other samples were very closely grouped. In Set (2) the Ni₂O₃ sample contracted most and the Fe₂O₃ sample least when compared to the standard without additive for this set. The temperature of purification did not appear to make any difference. In Set (3) the Ni₂O₃ sample contracted slightly more than the Fe₂O₃ and standard samples.

The coefficient of thermal expansion did not show any marked changes, within each set, either in the unirradiated or first-exposure data.

Argon-Sensitized Radiolysis of Carbon Monoxide - Water-Vapor Mixtures. The helium sensitized radiolysis of carbon monoxide and water-vapor mixtures was reported previously. The data showed that all of the energy absorbed in the helium was effective in causing the carbon monoxide - water-vapor reaction even when the gas contained more than 99% helium.

Since argon has a lower ionization potential than helium, it is of interest to investigate the efficiency of energy transfer from the argon to carbon monoxide and water vapor. We have therefore studied

the radiolysis of carbon monoxide - water vapor mixtures in varying quantities of argon. The results are summarized below.

Sample No.	% A	% CO	% H ₂ O	$\frac{G(H_2)}{\text{Molecules/100 ev}}$	$\frac{G(CO_2)}{\text{Molecules/100 ev}}$	Total Pressure, torr
35-R-6	24.8	38.2	37.0	3.14	5.72	704
36-R-6	55.8	21.7	22.4	3.20	4.42	704
37-R-6	75.4	12.6	12.0	3.63	5.98	706
38-R-6	85.7	6.97	7.36	3.49	4.51	717
39-R-6	90.2	4.95	4.88	3.86	5.98	706
40-R-6	93.9	2.94	3.17	3.91	5.01	582

The data indicate that energy absorbed in the argon is also effective in causing reaction in all gaseous mixtures studied. Further work is currently in progress to extend the data to mixtures containing as much as 99.5% argon.

Oxidation of Boronated Graphite. As reported earlier, oxidation of a graphite sample containing a nominal 7 wt% boron (grey material, resulted in a rapidly decreasing oxidation rate, presumably as a result of the formation of B₂O₃, which serves as a physical barrier to further oxidation. To determine the effect on the oxidation rate of removing this B₂O₃, the same sample was placed in a container of distilled water, a vacuum applied to the container to remove air from the pores of the sample, and then the vacuum released to force water into the partially evacuated pores. Next, the water was boiled to dissolve as much B₂O₃ as possible, the sample removed from the water and out-gassed to determine the weight of B₂O₃ lost, and the oxidation rate was determined. After the rate measurements, the entire procedure was repeated. In all cases oxidation was carried out at 730 C with helium containing 1% O₂ at a flow rate of 150 cm³/min. The results of these treatments are presented in the following table:

Run	Wt. of B ₂ O ₃ Removed, g	% of Initial B Removed	% Oxidn. of Sample	Initial Oxidn. rate, hr ⁻¹	Final Oxidn. rate, hr ⁻¹	$\frac{-\Delta n_c}{\Delta n_{B_2O_3}}$
1	0	0	--	--	-2.53×10^{-4}	12.6
2	0.0133	10.1	6.07	-1.51×10^{-2}	-3.32×10^{-3}	7.18
3	0.0151	21.4	10.1	-2.54×10^{-2}	-6.54×10^{-3}	8.66
4	0.0156	33.2	15.1	-2.30×10^{-2}	-7.36×10^{-3}	6.87
5	0.0286	55.0	22.4	--	--	--

The values in columns 2 through 5 refer to the start of the run. A comparison of the initial oxidation rates of column 5 to the final oxidation rate of the preceding runs, column 6, demonstrates that B_2O_3 removal effects an appreciable rate increase. In contrast, when the initial and final rates for a given run are compared, it is seen that a significant decrease in rate occurs with the formation of B_2O_3 . The ratio of the moles of carbon lost to the moles of B_2O_3 formed, column 7, gives an indication of the relative rates of graphite and boron oxidation. The general decrease in this ratio may indicate an increasing accessibility of the boron to oxidation as a consequence of graphite removal, or a more effective inhibition of graphite oxidation by the remaining boron. Unfortunately, the fragile sample was accidentally broken after the leaching treatment of Run 5.

Pore-size-distribution measurements have been performed on all four types of boronated graphite but without the success ordinarily found with other graphites. For an unknown reason, the specific surface areas obtained by summing the pore areas are larger than the specific areas found by the BET method, the discrepancy being greatest for the samples containing a nominal 7 wt% boron. This may be seen in the following table.

<u>Sample</u>	<u>Boron Content</u>	<u>BET Surface Area,</u> <u>m²/g</u>	<u>BJH Surface Area,</u> <u>m²/g</u>
D-80-T	5% (black)	0.46	0.48
E-78-T	5% (grey)	0.15	0.14
H-79-T	7% (black)	0.30	0.34
J-70-T	7% (grey)	0.20	0.25, 0.24

Qualitatively the pore-size-distributions are similar to those found for other graphites, with a peak in the 20-25 Å pore-radius range and usually a secondary peak or peaks at larger radii. The significant difference in the surface areas of the black and grey materials results from the greater number or pores of all sizes in the micro-pore range for the black boronated graphite.

7. Metallic Fuel Development

Irradiation of Thorium-Uranium Fuel Elements. The irradiation of six tubular Zircaloy-2 clad thorium-2.5 wt% uranium-1.0 wt% zirconium fuel elements continued successfully in the ETR-P7 and M-3 loops. Current irradiation conditions are:

<u>Loop</u>	<u>Core Temp., °C</u>	<u>Surface Heat Flux BTU/hr-ft²</u>	<u>Integrated Exp. Fissions/cm³ (Mwd/ton)</u>	<u>Fuel Volume Increase %ΔV/V</u>
P-7	425	5.1×10^5	3.9×10^{20} (11,100)	1.0
M-3	520	7.1×10^5	2.1×10^{19} (880)	-

Irradiation of Metallic Uranium with Submicron Uranium Carbide Dispersions. The irradiation performance of uranium containing a submicron dispersion of uranium carbide is being evaluated.

Density measurements have been completed on four fuel rods, three with 0.5μ diameter UC particle dispersions, and one with UC particles in the size range 2-5μ, which were irradiated to 0.3% burnup at a maximum fuel temperature of 500 C. The density decrease of the fine UC containing fuel rods, all of which suffered cladding failures, ranged from 8.8% to 13.4% whereas the decrease for the fuel rod with larger UC particles was only 3.4%.

Metallographic examination of the uranium in the rod with the larger UC particles shows considerable porosity of about 1μ size with some small amount in the 10μ range. Most of this porosity is spherical. In the fuel with the submicron UC dispersion the uranium exhibits extensive grain boundary tearing porosity. The difference in the as-irradiated densities is readily confirmed by the large difference in size and amount of porosity observed.

Postirradiation annealing of declad samples at 625 C for 10 hours has produced approximately twice the density decrease in the uranium with the 2-5μ particles as in the uranium with the submicron UC. The density changes in these rods follow the same trend as was observed in similar samples examined earlier.

8. USAEC-AECL Cooperative Program on Development of Heavy Water Moderated Power Reactors

Work was started on installing the pressure tube and preheater in the heat transfer laboratory for running the fog-cooled experiments. This work requires the removal of the existing preheater, installation of a new preheater to generate 100% steam, installation of the AECL flow nozzle, and installation of the connecting piping. Simultaneously, assembly of the 19 rods for the electrically heated test section was continued. This test section consists of 6.5-ft long rods spaced 0.040 inch apart in a 3.25-inch pressure tube. The test section is designed for a maximum heat flux of 680,000 Btu/hr-ft².

The tests have been designed to cover both fog (wet steam) and sub-cooled water inlet conditions. For fog-cooled conditions the tests will compare the differences in boiling burnout heat fluxes when using the AECL nozzle. This nozzle can be adjusted to provide a homogeneous fog, or a separated fog flow, at the test section inlet. The separated fog flow will have the water concentrated at the interior of the bundle. The tests will also determine the effects on boiling burnout of a smooth versus a roughened pressure tube wall. The rough wall is created by 0.050-inch high circumferential ridges every 3.25-inch along the length of the tube.

9. Advanced Reactor Concept Studies

Nitride Fuel Cycle Studies. Document BNCC-43, "Economic Considerations for a Nitride Fuel Cycle Study," was issued during the month. This document reviews and evaluates the fuel cycle costs reported in a Combustion Engineering, Inc., study of a carbide-fueled 1000 Mwe fast breeder reactor⁽¹⁾, with comparisons to the expected costs of a nitride fuel cycle. These comparisons, included in the previous monthly report, indicated that the costs of nitride fuel conversion and fabrication may be critical to the cycle, and that any development of such a cycle should include efforts toward development of an economical process for conversion of fuel materials to the nitrides.

Cross section data for the 18-group library to be used in the nitride fueled fast reactor study have been transferred to cards in HFN input format. This was accomplished using a new version of HRG developed by Theoretical Physics which now provides for punching microscopic cross sections in addition to the macroscopic punch option, formerly available. A detailed examination of the library data is being carried out to be certain that best available values are utilized before generating the HFN library tape.

Gas generation and transport in mononitride fuels has been briefly reviewed. Although there appears to be only limited experimental data on fission gas release from nitrides currently available, similar data for carbides are more abundant. Both uranium monocarbide and mononitride have similar crystal structure (FCC) and lattice dimensions that are nearly the same. On this basis one might expect the diffusion properties of noble gases in nitrides to be somewhat similar. Interestingly, diffusion measurements of xenon in UC and UN yield essentially the same activation energy of about 60 k Cal/mole. Calculation of gas generation rates in the fast neutron spectrum from all sources in the fuel was initiated.

(1) Liquid Metal Fast Breeder Reactor Design Study, Combustion Engineering, Inc., CEND-200, Vols I and II.

10. Critical Flow of Steam-Water Mixtures

The high pressure critical flow apparatus was readied for critical flow experiments using a "saturated" blowdown of heated water. This method differs from previous tests which used high pressure air to maintain the pressure in the water storage tank at a constant pressure.

The first experiment used the test section employed earlier and reported in HW-80535. This test section was in the form of a pipe 0.500-inch ID and 10 inches long. Pressure taps were provided at close intervals along the length of the pipe to enable the determination of axial pressure gradients during the blowdown of the 1000-gal vessel. The purpose of this experiment was to provide data to allow a comparison of the results of transient experimentation with those of the steady state method used earlier. The water in the vessel was heated to 535 F (280 C) (saturation pressure = 920 psia) and the experiment initiated by the impulse opening of the main discharge valve, thus allowing the heated water to pass through the test section to the atmosphere. During the discharge of the water in the vessel (which took approximately 12 minutes), the pressures at the various positions in the test section and in the tank were measured by transducers and recorded on an oscillograph. Flow rates were measured both by drop of water level in the vessel and by an orifice flowmeter. The test was successful, and the resulting data were prepared for further processing in the computer center.

11. Upstream Boiling Burnout

Additional experiments were run to investigate conditions at which boiling burnout takes place at locations upstream of the outlet end of uniformly heated test sections. The test section used consisted of a 12-ft long electrically heated tube with flow through a 0.435-inch hole. The test section was constructed so that it was possible to heat the entire 12-ft length, the upstream 6-ft alone, or only the downstream 6-ft length.

Extensive tests were made at 1500 psig with a flow rate of 7×10^6 lb/hr-ft². The following are the most significant observations of these tests:

- a. Burnout was observed at locations ranging from the downstream end to 14 inches from the upstream end of the heated section. In 15 out of the 24 tests, burnout occurred at a location away from the downstream end.
- b. Although the inlet enthalpy varied from 400 to 700 Btu/lb and the outlet enthalpy from 630 to 850 Btu/lb, the

enthalpy at the point of burnout fell in a much narrower band of 630 to 730 Btu/lb (3-12% quality). As the inlet (or outlet) enthalpy was raised, the location of burnout moved further upstream, thus producing only a small increase in burnout enthalpy.

- c. With all three modes of heating, the burnout heat flux decreased with increasing enthalpy at the lower enthalpies where downstream burnout occurred. At an outlet enthalpy of about 675 Btu/lb (11% quality) the burnout started to shift to upstream locations and the burnout heat flux reached a minimum of about 1.05×10^6 Btu/hr-ft². With 12 feet heated, the burnout heat flux remained at this value on further increases of enthalpy. However, with the downstream 6 feet heated, the burnout heat flux rose again with increasing enthalpy to 1.6×10^6 Btu/hr-ft² for 730 Btu/lb at the upstream burnout location.
- d. In those runs where downstream burnout was obtained (i.e., those at lower inlet enthalpies), the burnout heat flux was 10-30% higher (at the same outlet enthalpy) with the upstream 6 feet heated than with the entire 12 feet heated. With the downstream 6 feet heated, the results were intermediate.
- e. In two identical runs with 12 feet heated, burnout occurred upstream of the center. In two other runs with the upstream 6 feet heated and the same inlet enthalpy (600 Btu/lb), burnout was observed at the same location and at the same heat flux.
- f. Three runs were repeated to check reproducibility. Agreement was excellent with respect to both burnout heat flux and position.
- g. The results, with 12 feet heated, agreed well with previous data obtained with a 12-ft test section. The present test section differs from the old only in having axial inlet and outlet flow and having 18-inch unheated lengths at the inlet and outlet rather than radial inlet and outlet flow at the ends of the heated section. (All these burnouts occurred at least 4 feet from the start of the heated section.)
- h. Measurements of pressure fluctuations in the test section were made with a high speed recorder to determine whether such fluctuations were related to upstream burnout, as seems to have been the case in low pressure tests. The magnitude of

the pressure fluctuations was comparatively small, on the order of 10 psi, and preliminary examination of the data revealed no relation to burnout.

Although any firm conclusions must await more complete analysis of the test data and the completion of additional tests, two conclusions seem to be indicated by the results of these and previous tests of upstream burnout at high pressure:

- a. The graph of burnout heat flux versus local enthalpy (for the diameter, pressure, and flow used in these tests) passes through a local minimum.
- b. The burnout heat flux is higher at locations near the entrance to the test section or the start of the heated length than when the same local enthalpy exists further downstream.

12. Plutonium and U-233 Fueling of a Fast Compact Reactor

Work on this program has been completed except for completion of the final reports. The final report of the study is approximately 60% complete in rough draft form. Rough draft documentation of the core thermal hydraulic code, SWISSCHEESE, was completed. A document describing the gas transport property computer program, MIX, is being prepared for issuance in final form.

13. Fast Flux Test Facility Design Studies

Important progress items during the month were (a) review and acceptance of the FY-1965 study plan by AEC Headquarters, (b) extensive travel by FFTF personnel to obtain current data on sodium technology applications in fast and thermal reactor service, (c) initiation of use of six off-site firms and consultants, and (d) need for early initiation of irradiation experiments to prove out the suitability of PuO_2 cermet as driver fuel for the FFTF.

A draft of the Progress Report, Fast Flux Test Facility Studies (April 1964-April 1965), was prepared and is currently under management review.

A \$20,000 Procurement Directive (RLOO-65-1) was placed with Atomic International for assistance in conceptual design of the FFTF materials handling system.

A \$20,000 Procurement Directive (RLOO-6502) was placed with Battelle Memorial Institute, Columbus, Ohio, for assistance in conceptual design studies.

Consultant Agreement BCA-48 was placed with Gardner L. Locke, with a maximum limit of \$5,000.

At month-end procurement activities were in progress to obtain \$15,000 of service from Argonne National Laboratory and \$30,000 of service from Atomic Power Development Associates. Arrangements are being made to obtain, under an existing USAEC-COO contract, about seven man-weeks (\$7,000) of service from Mr. J. DeFelice, President of Nuclear Technology Corporation.

With the cooperation of the financial representative, a special cost accounting and weekly cost reporting system was set up. Costs are coupled directly to the individual principal work assignments in the program. This is the initiation of a procedure which will be expanded to a full PERT-cost system.

Preliminary estimates of the PACE-funded design costs for FFTF were made as follows:

\$ 600,000 - FY-1966	Advance Engineering
\$1,000,000 - FY-1967	\$500,000 Nuclear Engineering \$500,000 Title I Design
\$2,200,000 - FY-1968	\$500,000 Nuclear Engineering \$1,700,000 Title II Design.

Revised procedures for construction budget reviews were issued by the AEC January 5, 1965, but did not arrive at BNW until early April. This was too late to incorporate the effect of the new procedures into the current \$75,000,000 FFTF cost estimate.

Review of the procedures indicated that the FFTF estimates for contingency (16.7%) and escalation (8.5%) may be low. The revised procedures would indicate 18% contingency and 17-1/2% escalation and would result in increasing the FFTF cost estimate from \$75,000,000 to \$82,500,000.

The FFTF design estimate (26.3%) is consistent with the range of 12-32% costs shown in the AEC procedures and is slightly (but justifiably) higher than the recommended 20% for complex projects.

Existing data from research performed for Fermi on irradiation effects in boronated graphite have been compiled in memorandum form for FFTF use. These data include mechanical, thermal, and physical property changes resulting from irradiation in Hanford facilities at 370 and 480 C to exposures of 5×10^{21} n/cm²-sec (thermal).

Several studies are in progress on various aspects of the reactor cooling systems. Included are studies for establishing (1) the number

of parallel loops, (2) the method of dumping reactor heat, and (3) a method for process control of the primary, secondary, and tertiary cooling systems. In addition, a machine program is being prepared for analyses of hydraulic and thermal transients caused by simple loop perturbations, loss of pump power, and scrams. This program is needed before the method of process control can be firmed up and before a satisfactory emergency cooling concept can be developed.

A review of the relative merits of Na or NaK for the secondary coolant system has been completed. While NaK is liquid at room temperature, there are a number of more important factors which appear to be in favor of sodium, and a decision has been made to use Na as the secondary coolant.

An exhaustive study is being made of candidate driver fuel materials, in particular the cermet type, as requested by the USAEC at the FFTF meeting in Germantown, Maryland, in April 1965. A large amount of unclassified work has been conducted in the USA on cermet fuel development, with particular emphasis on the UO_2 -stainless steel type. Experimental work on UO_2 - and $(\text{U,Pu})\text{O}_2$ -stainless steel cermet pins and plates was carried on in the U.K. up until about two years ago when the work was discontinued because of poor performance of the high ceramic loading (to 50 vol%) type fuel and a breeding penalty.

A metal fuel of interest to the FFTF is Nb-20 wt% U alloy. Fuel performance can be termed highly promising on the basis that most of the density decreases observed (2.5-3.0% of total atom burnup) are required to accommodate the fission products. Bare, unrestrained pins irradiated in NaK, with maximum fuel temperatures to 1040 C, and to total atom burnups to 0.9% or U atom burnups to 9.7% showed no evidence of poor performance or interaction with the coolant.

14. Nuclear Rocket Fuels Studies

Studies conducted for AEC-SNPO on tungsten- UO_2 fuel element development and on fueled graphite problems are reported via a separate distribution list.

D. DIVISION OF RESEARCH - 05 PROGRAM

1. Radiation Effects on Metals

This program is directed toward establishing the combined effect of impurities and neutron irradiation on the properties and structure of specific metals, and deducing from thermally activated recovery processes how the damage state can be altered. Present studies involve single and polycrystalline specimens of molybdenum, nickel, and rhenium.

Molybdenum foils irradiated to 10^{20} nvt have been annealed at 575 C and examined by transmission electron microscopy. The only significant difference between this microstructure and those at lower exposures was the "smoothing out" of the dislocation lines and the appearance of more circular type loops. At these temperatures, only those processes requiring relatively short range dislocation motion (e.g., climb) are occurring.

Investigations of the nature of the loops formed in molybdenum irradiated to 10^{20} nvt and annealed at 750 C or higher are being continued. On relatively few measurements has it been possible to obtain ideal diffracting conditions. Nevertheless, some vacancy type loops have been identified. Interstitial loops are present in much greater number. At the present time, it is not clear as to why there should be so few vacancy loops. Apparently, most of the free vacancies and small vacancy clusters have been annihilated at an early stage and only the largest vacancy clusters are stable enough to form loops.

The role of dislocations as sinks for the point defects created during irradiation is currently being studied by irradiating pre-strained samples. Transmission electron microscopy shows no depletion of the spot defects around dislocations in the irradiated samples which had been pre-strained up to 20%. An actual count of the number of spot defects revealed no difference between pre-strained and annealed samples. In this case, it is most likely that the migration distance of the point defects at the irradiation temperature is so short that any denuded zone about a dislocation would be of the same magnitude as the distance between the spot defects. These pre-strained and irradiated samples will be subsequently deformed in order to study the effect of irradiation on dislocation sources. In particular, it will be noted whether the dislocations formed during the pre-strain serve as sources after irradiation or whether new sources such as grain boundaries are activated.

Rhenium samples irradiated to 7×10^{18} nvt have been further annealed for two hours at 800 C and at 1000 C. At 800 C, there is a slight coarsening of the spots and a decrease in the spot density from 4×10^{16} to $1.5 \times 10^{16}/\text{cc}$. Upon annealing at 1000 C, there is no further coarsening but a large decrease in the spot density, from 4×10^{16} to $1 \times 10^{15}/\text{cc}$. This behavior is very similar to the annealing behavior of the irradiated JM molybdenum in which there was some coarsening and then disappearance of the spots but no loop growth. Rhenium samples are currently being irradiated to doses of 1.5 and 3×10^{19} nvt. It is hoped that larger spots and loops will result and that the character of the defects can be determined.

Low temperature annealing experiments on irradiated (1×10^{19} nvt, $E > 1$ Mev) molybdenum foils have shown that there is no measurable

decrease in lattice parameter in high purity foils annealed at 200 C. There is a small but measurable decrease, 0.003 to 0.005%, for foils containing 100-200 ppm and 400-500 ppm carbon.

Annealed single crystals of molybdenum have been prepared with pairs of faces cut parallel to crystal planes having the same spacing but different crystal directions. Pairs chosen are (330), (411); (550), (710); (550), (345); and (600), (442). Eight crystals in all have been prepared. These crystals will be used in x-ray diffraction experiments to determine the existence of crystallographically-oriented defect configurations. After complete pre-irradiation characterization, they will be submitted for irradiation.

A capsule containing single-crystal length-change and x-ray specimens has been opened after reaching a goal exposure of 1×10^{20} nvt ($E > 1$ Mev). Examination of the specimens is not complete, but preliminary measurements show the magnitude of the length increases to be greater the lower the carbon content of the crystal.

X-ray diffraction determinations of lattice parameters are in progress.

The temperature-dependence of the upper and lower yield stresses in irradiated and unirradiated molybdenum has been found to contradict the dependence predicted by existing theories of yielding based on the concept of the breaking away of a dislocation from a solute atom atmosphere. This lends further support to the conclusion that yielding in molybdenum is controlled by the rapid multiplication of dislocations. Calculations of the degree of multiplication during yielding have shown that a two-fold increase in the number of mobile dislocations occurs between the upper and lower yield points in unirradiated molybdenum. The extent of multiplication in irradiated molybdenum is much greater, leading in some cases to as high as 20-fold increases in mobile dislocation density.

2. Plutonium Physical Metallurgy

The objective of this program is the derivation of fundamental information relative to (1) the kinetics and mechanics of the phase transformation in plutonium, and (2) the mechanisms by which monoclinic plutonium deforms.

As has been reported in the past, the amount of evidence for the existence of definite crystallographic relationships between the alpha and beta phases continues to increase. An effort is currently under way to establish, if possible, the details of this relationship.

The study of the texture established by the transformation from beta to alpha under the influence of a compressive stress offers the most promising approach to this problem. Through the use of the pole figure goniometer it is possible to find regions which behave from a diffraction standpoint essentially as a single crystal.

A technique has been worked out which permits the goniometer mounted sample to be increased sufficiently high in temperature to effect the alpha to beta transformation. With proper attention given to the glovebox atmosphere the surface oxidation can be minimized to an inconsequential level for periods of time sufficient to thoroughly investigate the beta diffraction pattern as well as that of the alpha resulting from a subsequent transformation. These pseudo-single crystal regions can thus be located in the alpha phase and the behavior of a given reflection followed through the transformation and the resulting beta structure related to the orientation of the parent alpha. The effects of interphase cycling on the orientation of both phases will also be given attention.

Samples have been prepared and textured which will be subjected to tensile loading in the hope of producing slip lines in crystal surfaces, the orientation of which is known.

An explanation for the formation of disks of columnar alpha grains has been formulated. The disks appear as bands on polished and etched surfaces and they are particularly evident in plutonium that has been reheated into the beta phase and transformed at high alpha temperatures. They are responsible for the macroscopic veining frequently observed on this surface of plutonium.

The formation of the large disk-shaped volumes with their columnar grains is reminiscent of the internally twinned martensitic plates that form in iron alloys; however, they differ in that they are much larger and that their component alpha grains appear to be columnar crystals and not simply twins.

In order for a disk-shaped transformed region to grow in thickness while maintaining continuity with the parent phase across the approximately planar interphases, the volume change must be accommodated almost entirely by expansion or contraction perpendicular to the plane of the disk. Thus, for a disk, $\Delta L/L_0 = V/V_0$ in the direction perpendicular to the flat interface and $\Delta L/L_0 = 0$ in the plane of the interface. The growth of individual grains in disk shapes is common during transformation. The alpha plutonium disks are unusual because they are polycrystalline.

Experimental evidence indicates that the mode of beta to alpha transformation is martensitic. The first grain or nucleus of alpha to form from the surrounding beta parent phase would undergo an anisotropic shape change. This would result in an anisotropic stress field in the beta matrix near the alpha nucleus. Subsequent nucleation would be favored by the stress in some regions of the stress field and inhibited in others. Also, the orientation of subsequent nuclei would be influenced by the stress field of the first nucleus; those nuclei whose shape change was in the direction to relieve the stress would be favored. The disk shape of the transformed region suggests that the stress field surrounding the first nucleus may have cylindrical symmetries; secondary nucleation is favored in a plane. Thus the edgewise growth of the transformed region would be a continuing process of sympathetic nucleation resulting in a polycrystalline disk of increasing diameter. The growth of the individual alpha grains in the disk would be limited by impingement with their neighbors to growth in the direction perpendicular to the plane of the disk.

E. CUSTOMER WORK

1. Radiometallurgy Laboratory

Examinations. The results of routine examinations and measurements are or will be reported as part of the sponsoring research and development programs. During the month, examination and testing included the following:

Photomicrography - - - - -	59
Photomacrography - - - - -	62
Photomosaics - - - - -	9
Macrohardness - - - - -	93
Microsampling - - - - -	10
Tensile Testing (Rm Temp)- - -	17
Tensile TEsting (Hi Temp)- - -	94
Replication - - - - -	12
Autoradiography - - - - -	20
Decladding - - - - -	41
Dissolution - - - - -	6
Density - - - - -	40
X-ray - - - - -	6
H ₂ Analysis - - - - -	27
Anneal - - - - -	2
Fission Gas - - - - -	4

Replication of Irradiated Components by Molding. Evaluation of candidate molding materials was started to improve the dimensional accuracy of replicas made by remote molding techniques. A new casting resin, "Epoxical" thick-section casting resin No. C-301, was tested. The

dimensional accuracy, reproducibility, and mold release characteristics of this resin were excellent. The resin has a low viscosity (7,000 cps), a working time of 45 to 60 minutes, and low shrinkage.

Stereo Zoom Microscope. The positioning motor for the stereo zoom microscope slide has been modified for either DC stepping or AC operation. This change permits the microscope to be used for remote measuring as well as viewing. As-built drawings of the microscope positioner and control circuits are being prepared.

2. Metallography Laboratories

Routine metallography work will be reported as part of the sponsoring research and development component's work. However, items of unusual interest or representing departures from routine operations will be reported here.

Two samples of montmorillinite have been examined to establish that the electron microscope can be used as a tool for particle size and shape determination of this material. A dilute, aqueous solution of the samples was made and a drop deposited on an SiO covered screen and allowed to dry. The particles are somewhat sensitive to the electron beam but with proper care can be examined and photographed with no difficulty. Many of the clay particles are thin enough to allow transmission microscopy, and electron diffraction patterns can be taken with no damage to the material.

A series of zirconium - 5% beryllium samples with various heat treatments were submitted for metal-quality examination. As a sidelight to the routine examination, the effect on the morphology of this alloy was observed. The morphology, or shape change during solidification, paralleled that of AlSi, a system which has been known for some time. A quick chill from the melt produces a highly acicular pattern of $ZrBe_2$ crystals with zirconium between them. Slow cooling from the melt produces square chunks or cubes of $ZrBe_2$ with large areas of pure zirconium between.

During the month, 726 samples were processed, 261 electron micrographs and a total of 1401 optical micrographs and macrographs taken, 2600 negatives printed, and 11,392 prints processed.

3. High Temperature Lattice Test Reactor (HTLTR)

HTLTR Mockup. Run No. 2 goal core temperature of 700 C was reached and maintained for a 115-hour period. Subsequent to this, the mockup was cooled down for heater element and control and safety rod inspection.

Results of B₄C and graphite samples which were intermittently inserted in the core as follows:

B₄C Samples

Sample No.	1	2	2	2	2	2
Exposure time, hr	91.8	24.2	17.7	2.2	14.7	6
Core Temp, °F	520-1020	1020-1070	1260-1310	1310	1300	1300
Initial Wt, gm	12.1733	11.9005	11.8952	11.8986	11.8977	11.9006
Final wt, gm	12.1687	11.8952	11.8986	11.8977	11.9006	11.9008
Wt Change, gm	-0.0046	-0.0053	+0.0034	-0.0009	+0.0029	+0.0002

Graphite Samples

Sample No.	14	14
Exposure time, hr	24.8	64
Core Temp, °F	1070-1260	1290
Initial Wt, gm	13.6483	13.6438
Final Wt, gm	13.6446	13.6429
Wt Change, gm	-0.0037	-0.0009

The 13 graphite samples which remained in the mockup for the duration of Run No. 2 had weight losses ranging from 0.0010 to 0.0047 gram with an average loss of 0.0030 gram.

The 2-1/2" x 1-1/16" x 3/8" B₄C sample placed between two TD nickel plates exhibited a weight gain of 0.0881 gm. Initial and final weights of the B₄C sample were 28.2720 gm and 28.3601 gm, respectively.

The mockup was inspected after Run No. 2, and no increase of core corrosion over that observed at the end of Run No. 1 was visible. One heater element was removed, and no malfunction or corrosion could be detected.

HTLTR Safety and Control Rods. The following is a resume of the problems encountered in the first hot test of the rods:

Vertical Safety Rod: After significant heatup of the mockup core ($\sim 500^\circ\text{C}$), the rod stuck while pulling from the core. Each time the rod was pulled it stuck at between 70 and 80 inches (measured from its full in-position). Once stuck, the rod would neither scram nor pull any further. If left to cool for a long enough time to stabilize the temperature, the rod could be pulled or scrambled as if it had not been stuck. Sticking, temperature equalization, pulling, and successful scrambling of the rod occurred each rod operating cycle while the mockup was at temperature ($\sim 700^\circ\text{C}$). After cooldown no sticking occurred. Subsequent investigation indicated that the dummy spacer unit, installed outside the mockup shell below the bottom of the rod housing to allow full height drops of the rod, caused the sticking. Modification of the plate liner should correct the problem. The dummy spacer unit does not exist on the reactor so this problem may not be applicable to reactor operation.

Horizontal Control Rod: At temperature, the drive mechanism operated under much higher load during the last 1/10-inch of rod travel (inner end of travel). When under this high load, the scram latch would not release. Backing the rod out about 1/10 inch relieved the high load and the rod would scram satisfactorily. Disassembly of the control rod revealed no reason why the heavy load should occur. It is possible that graphite chips, which were present, could lodge in the interface and cause the sticking. The chips were from the uranium oxide bearing graphite outer cylinders. The problem of the scram latch not releasing under high load had been investigated, and a modification has been added to the latch ring. This modification consists of providing a ramp for each ball which tends to make the ring unlock under increasing load on the latch. This arrangement will be tested during the next operating period.

Critical Mass Physics Support Work. Two concentrator boilup tests, using an $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ spiked solution as a standin for plutonium nitrate, were completed. The concentrator was filled with 200 g/l $\text{Al}(\text{NO}_3)_3$ solution to within approximately 3 inches of the top. Steam at a pressure of 60 psig was started to the coil and maintained at this pressure until the run was completed. Surging of the condensate was observed while the first four liters were condensed. After this, concentrator operation appeared to be normal.

Each liter of condensate collected was sampled and submitted for analysis. The first 3 liters collected averaged 13 g/l Al. Subsequent liters (4-10) dropped to 0.01 to 0.14 g/l.

Environmental Tests on HTLTR Materials. Two environmental tests were run to determine the effect of moisture in the HTLTR atmosphere on the corrosion of control mechanism material composed of 5% B_4C in a graphite matrix. These tests indicated that with water contents of 5000 ppm and 1800 ppm in the nitrogen atmosphere, corrosion of the

control material will occur at 1100 C. In order to obtain more quantitative data on the reactivity of B_4C with the reactor atmosphere, an exposure test was started involving pure, consolidated B_4C in nitrogen with 300 ppm H_2O at 1100 C. Periodic weight measurements of the specimen will be made.

4. EBWR Fuel Elements

Thermal Cycling of Aluminum Core EBWR Rods. The thermal cycling studies directed at determining possible dimensional changes in three aluminum-core EBWR rods have been completed. The rods were heated in a steam autoclave to 370 C and 40 atm in one hour, held at temperature of 3 hours, and cooled to 90 C and 1 atm over an 8-hour period. Two of the rods exhibited rod lengthening of approximately 39 mils after 6 cycles, but subsequent cycling induced rod shortening. After 48 cycles, one rod had shortened approximately 28 mils and the other approximately 63 mils. Anomalous results were obtained on one rod which exhibited little growth or shrinkage up to 28 cycles, but which suddenly started shrinking, leveling off at 48 cycles with a total decrease in length of 33 mils.

Irradiation Testing of EBWR Prototypic Fuel Rods. Seven capsules containing short lengths of vibrationally compacted, EBWR, UO_2 -1.5 wt% PuO_2 fuel rods are being irradiated in the MTR, and 25 have been discharged. The capsules continue to indicate satisfactory performance and have achieved maximum burnups of 4.7×10^{20} fissions/cm³ (16,700 Mwd/ton of fuel) and 4.8×10^{20} fissions/cm³ (17,000 Mwd/ton of fuel), respectively.

Fuel Element No. 6501, the 21-rod bundle (5x5 array with corner rods missing) with full-size replaceable EBWR fuel rods, is currently under irradiation in the core of the PRTR. Current burnup of the element is 1170 Mwd/ton of fuel or 0.33×10^{20} fissions/cm³. It is tentatively planned to remove and replace one rod when the burnup reaches $(0.4-0.6 \times 10^{20})$ fissions/cm³ (1500-2000 Mwd/ton of fuel).

Continued satisfactory behavior has been noted with the twelve, 3-rod clusters of full-size (148-cm long) EBWR rods under irradiation.

5. Other Customer Work

Pu-240 and Pu-241 Irradiation Samples for Phillips Petroleum.

Twenty-three samples of Pu-240-Al and Pu-241-Al alloys are required for fission product transient measurements in the MTR by Phillips Petroleum Company. These samples are similar to samples fabricated in 1962 and 1964 of Pu-239, U-235, U-233, Au and Li aluminum alloys. The samples are tubular and made by coextrusion cladding. The cladding is a high purity aluminum - 1.15 wt% Si (for hardening) alloy. The Pu-240-Al alloys will be made in five concentrations up to

1.6% Pu and the Pu-241 alloys will be made in five concentrations up to 3.2% Pu.

Because of the relatively short half life of Pu-241, the samples will be fabricated as rapidly as possible. In order to permit more rapid fabrication, a die pressing technique which will reduce the machining and handling time is being developed to pre-shape the plutonium alloys prior to extrusion. The cladding alloys have been cast and extruded. The cladding components, tooling, and molds are being fabricated. The samples are scheduled for fabrication in June.

Al-Pu Discs for the Phoenix Program. Five kilograms of plutonium are being fabricated into discs for the Phoenix program. The discs are similar to those fabricated in 1964 by the "spiral-machining" process. The fabrication process consists of casting aluminum 20 wt% Pu alloy billets, extruding the billets into bars, machining the bars into a spiral turning, cutting discs from the turnings, and covering each disc to final size. Four kilograms of plutonium were alloyed and cast into billets to start the fabrication.

IPD PCTR Al-Pu Alloy Fuels. Sixty fuel rods are to be fabricated for this test. Three groups, 15 rods each, will have plutonium-aluminum alloy cores. The alloys will contain approximately 2%, 6%, and 15% plutonium. The fourth group of fuel will be fabricated from compacted thoria. All the fuel will be clad in aluminum. The elements in each group range in length from approximately 7 inches to 35 inches. It is planned that the 6% alloy fuels will be completed by May 1, 1965, followed by the thoria early in May and the 15% and 2% fuels later in the month.

Pneumatic Impaction of UO₂. Work was completed on three contracts for pneumatic impaction of enriched UO₂ for off-site customers. A total of 27 impaction shots were required, in both 2-1/2-inch diameter and 4-inch diameter sizes. Fifty-seven kilograms of enriched UO₂ was impacted, comprising eight different levels of enrichment ranging in U-235 content from 10% to 50%. This work was performed at the request of the Atomic Energy Commission.

HTLTR Fuel Development. A method was developed to weld TD nickel sheet of 20-mil thickness. This magnetic force welding method provided samples for use in evaluation of weld quality by metallographic examination and tensile testing, and for corrosion resistance testing. Metallographic examination and tensile testing indicate magnetic force welding to be a very positive process for joining TD nickel. The tensile test results indicate that room temperature strength at least 70% of that of the parent material can be readily achieved.

UO₃ Characterization. Characterization of UO₃ from the Chemical Processing Department was begun. The UO₃ which was reduced before ballmilling appears to have better sintering characteristics than the material which was reduced after ballmilling. Pellets which were sintered at 1650 C for an 8-hour period attained approximately 90% theoretical density. The O:U ratio of the presintered material was 2.018.

Isotope Source Fabrication. Preparatory development work for the fabrication of P-32, Pr-143, and Ca-45 isotopic disc sources, in aluminum and lead matrix material, was completed. An aluminum disc and a lead disc (3-inch diameter x 1/4-inch thick) were fabricated to evaluate cold pressing parameters and determine acceptability to the customer prior to dispersion of the isotope in the matrix. Fabrication of six disc sources for Radiological Physics, Physics and Instruments Department is scheduled for completion during May 1965.

Saxton Fuel Element Fabrication. Sixty-eight Zircaloy-4 clad Saxton rods were fabricated by vibrational compaction. Densities varied from 86-88% TD. An additional 20 rods will be fabricated by the end of the month. These will include five Zircaloy-4 clad removable rods for a 9 x 9 cluster and 10 rods clad in 15-mil stainless steel. This will complete the Saxton fabrication. Final shipment will be made on May 5, 1965.

Defect Propagation in Zr-4 Cladding. In order to establish the effect of manufacturing defects on fuel rod performance, a study was initiated to determine the effect of thermal cycling on tubing containing rejectable size internal surface defects. Three fuel rods were fabricated by vibratory compaction using Saxton tubing rejected at Battelle-Northwest because of defects in excess of 0.001 inch. The rods are heated in a steam autoclave to 400 C and 68 atm in one hour, held at temperature for three hours, and cooled to 100 C and 1 atm over an 8-hour period. After a thermal cycle, the rods were ultrasonically inspected in region of the known defects. Two rods remained unchanged, but examination of the other indicated propagation of three defects present in the cladding had occurred. Cycling will be continued until definite trends are established, after which time, metallographic examination will be performed on the defects.

Boiling Burnout Studies for the ATR. Heat transfer experiments were conducted with an electrically heated test section which is a full length simulation of a typical rectangular coolant channel of the Advanced Test Reactor. The test section coolant channel was 2.00" wide by 0.070-inch thick by 49.5 inches long. Both sides of the test section were heated along a 48-inch length by passing direct

direct current through copper-nickel bars which were brazed to the 0.010-inch thick stainless steel forming the flow channel. The Cu-Ni bars were machined in an axial profile along the back side to achieve an axial, chopped-cosine heat input pattern with a peak-to-average value of 1.4:1.

The experiments conducted to date were principally the determination of hydraulic demand curves at constant power input rates with coolant velocities from about 1.5 to 5 and 25 to 50 ft/sec. The lower velocity range represents reactor shutdown or emergency conditions while the higher range is typical of normal reactor operating conditions. These experiments were conducted by establishing coolant inlet temperature, flow rate, and outlet pressure at a given power input rate. The pressure drop along the test section was then recorded as the flow rate was decreased in small steps, by an upstream throttle valve, with other parameters held constant. Flow reductions were continued until the channel pressure drop increased appreciably due to boiling and two-phase flow conditions.

The experimental results show that in general the hydraulic demand curve minima occur at bulk coolant outlet temperatures which are slightly below the saturation temperature corresponding to the outlet plenum pressure. Flow rates, at the demand curve minima, are about 1.05 times the flow rates which would be calculated to achieve bulk saturation conditions. This undoubtedly results from nonuniform temperature distribution across the coolant channel and near the narrow edges of the coolant passage where the heat flux is relatively low. Special thermocouple probes were used to sense water temperatures near the outlet end of the coolant channel. The measurements show that temperature differences of 20 to 25 F between the channel center and the channel corners are common with an average "fuel plate" heat flux of 1.8×10^6 Btu/hr-ft² (which corresponds to an axial peak heat flux of 2.5×10^6 Btu/hr-ft²).

One experiment was conducted using a nearly flat supply curve to simulate the unstable flow conditions which result if the demand curve minimum exceeds the available or supplied pressure drop. For this experiment the power input was held constant at an average heat flux of 1.86×10^6 Btu/hr-ft² (2.6×10^6 Btu/hr-ft² peak) while the supply head was decreased in small steps. A flow path in parallel with the test section was used to simulate other fuel coolant channels and adjust the pressure drop supplied across the test section.

As the supplied pressure drop was reduced to slightly above the demand curve minimum, the flow rate became slightly unsteady and showed random fluctuations of about $\pm 3\%$. Then, as the supply

pressure was reduced further, the flow rate suddenly decreased at least 40%, and the heater bar temperatures rose very rapidly. By sudden interruption of electric power input, and by re-establishing the test section flow rate, damage to the test section was averted even though the heat flux at the middle of the test section was 2.6×10^6 Btu/hr-ft².

Manager
Reactor & Materials Technology

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