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REACTOR & MATERIALS TECHNOLOGY MONTHLY REPORT
JUNE 1965

F. W. Albaugh

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REACTOR AND MATERIAL TECHNOLOGY DEPARTMENT
MONTHLY REPORT - JUNE 1965

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

July 15, 1965

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REACTOR & MATERIALS TECHNOLOGY DEPARTMENT MONTHLY REPORTJUNE 1965TECHNICAL ACTIVITIESA. FISSIONABLE MATERIALS - 02 PROGRAM1. Metallic Fuel Development

N-Reactor Fuel Element Failures 2, 3, and 4. The second N-Reactor fuel failure which occurred May 15, 1965, has been examined in the Radio-metallurgy facility. Failure occurred through the inner clad of an outer component immediately adjacent to the contact area of a buggy-spring support. The site of initial water entry has been identified as a localized thin spot or defect in the clad. Detailed transverse metallographic examinations of sections through the area of the defect have not revealed any direct evidence that the defect was an inclusion, seam, lap, crack, or tear; however, the possibility of this type defect being the cause of failure cannot be ruled out at this time. Several features of the defect tend to support the possibility that failure resulted from fretting corrosion. Principal among these are the surface texture of the thinned area and its location relative to the support. Such a mechanism would depend on entrapment of foreign material by the support, with subsequent fretting between the foreign material and the cladding.

The third N-Reactor fuel failure occurred June 6, 1965, and was discharged without incident. Examination of the third failure in Radio-metallurgy revealed a circumferentially oriented pattern of blisters extending about 230° and involving contact areas of two of the three center inner component buggy-spring supports. The suspected initial site of water entry bears a striking resemblance in appearance and location to that of the second failure. Transverse metallographic examinations through the suspected site of water entry are being made to determine the nature of this site which, on macroscopic examination, appears to have been a small pit adjacent to one of the supports. Fretting corrosion rather than coextrusion or manufacturing processing seems to offer the best explanation of the source of the defect.

On June 15, 1965, a fourth N-Reactor fuel failure occurred in tube 1258. This tube was one of 38 used in a coproduct production test. The driver elements were heavy-walled single tubes of 1.25% enrichment. Greater than normal discharge forces were required to clear the column, apparently because the failed driver element was in contact with the process tube.

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Examination in Radiometallurgy indicates the failure occurred in the inner bore of the element with extensive fuel corrosion and undercutting of the clad occurring before clad fracture and gross fission product release occurred. The generalized nature of the damage is such as to preclude a reasonable estimate of the initial site of failure at this time. Detailed radiometallurgical examination is in progress.

Alternate Uranium Compositions. 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 treatments are being conducted in NaK capsules. Maximum uranium temperatures continue in the 600-625 C range in the peak flux capsules and in the 475-500 C range in the capsules at the ends of the columns. Estimated fuel exposures to date in center and end capsules of the columns are 2300 Mwd/ton and 1500 Mwd/ton, respectively. One column of eight capsules has been scheduled for discharge on June 30. The remaining four columns of capsules have a goal exposure of 3000-4000 Mwd/ton.

Irradiation of Metallic Uranium with Submicron Uranium Carbide Dispersion. The irradiation performance of uranium containing a submicron dispersion of uranium carbide is being evaluated. Metallography has been completed on samples which were postirradiation annealed at 625 C for 10 hours. Initial examination of the optical photomicrographs shows no significant change in the amount or size of porosity in either submicron UC containing uranium or the uranium with normal size UC. Electron microscope examination of replicas from the two samples has been started. Two samples, one from rod 32 and one from rod 2, have been postirradiation annealed in the beta phase at 710 C for 10 hours and density determination completed. During this anneal the submicron UC containing uranium increased in volume 1.55%, whereas the normal size UC containing uranium increased in volume 3.9%. These results are consistent with the volume changes measured on samples from the same rods that were postirradiation annealed at 625 C for 10 hours. The uranium with submicron UC does not swell as much as uranium with normal sized carbides at temperatures in the range where fission gases are the major contributor to uranium volume increases. After completion of metallography on the beta phase annealed samples, they will then be reannealed at 800 C.

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

Four Zr-2 canned specimens of irradiated Al-Li alloy containing either amorphous boron, boron carbide, gadolinium oxide, or an alloy of

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50 wt% Cu - 50 wt% Cd in a 0.125-inch diameter axial hole have been ruptured during ex-reactor heating tests. Radiometallurgy examination revealed that 50-60% of the Al-Li alloy was expelled from the Cu-Cd containing target and approximately 90-95% in the case of the Gd_2O_3 containing target. Metallographic examination did not reveal any Gd_2O_3 particles in the remaining Al-Li alloy. Examination is continuing on the Cu-Cd containing target. Reactivity measurements indicate that retained poison was approximately 96% in the Cu-Cd target and only 45% in the Gd_2O_3 target.

To determine how much of the buried or added poisons are retained in the Al-Li alloy after rupture, and how much is free and lost, pieces from the boron and B_4C containing targets are being measured for reactivity before and after cleaning and scrubbing.

Preparation and Properties of $Li_5AlO_4(5Li_2O \cdot Al_2O_3)$. The high lithia, lithium aluminate, Li_5AlO_4 , is attractive as a target material because of its high lithium content. The only known data for this compound is the x-ray diffraction pattern. (1,2)

Li_5AlO_4 was successfully prepared by heating an intimate mixture of lithium hydroxide and alumina in vacuum to 525 C. Formation was verified by x-ray diffraction. The reaction yield was quantitative.

A visual melting point determination was made using a platinum-rhodium thermocouple with its tip inserted into a sample of the powder. The powder was heated to 550 in vacuum and then to higher temperatures in a dry argon atmosphere at 25 psia pressure. The melting point was observed at 1120 ± 15 C. The fused material gave the same diffraction pattern as the powder except for several weak lines corresponding to the strongest peaks of $\gamma-LiAlO_2(Li_2O \cdot Al_2O_3)$.

Pellets, wrapped in platinum foil and placed in evacuated quartz ampules, were treated at 800, 1000, and 1100 C for 20 hours. There was considerable reaction of lithia vapors given off by the pellets with the quartz ampules. This means the compound has an appreciable lithia vapor pressure at and above 800 C. The pellet treated at 800 C swelled slightly. Pellets treated at 1000 and 1100 C slumped and partially, not totally, melted. This fact might indicate that the compound melts incongruently with the first liquid appearing between 800 and 1000 C. However, knowing that (1) ceramic compounds generally

(1) ASTM X-ray Powder Diffraction File.

(2) Lehmann and Hesselbarth, Z., Anorg. Allg. Chemie, Band 313, 117-120 (1961).

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have a very narrow composition range, (2) all three pellets lost considerable lithia, (3) the fused material had indications of gamma-LiAlO₂ (due to the loss of some lithia), one can logically propose that the compound melts congruently and that a eutectic exists in the Li₂O-Al₂O₃ system between the compounds 5Li₂O-Al₂O₃ and Li₂O·Al₂O₃ and between 800 and 1000 C.

These conclusions will be tested by x-ray diffraction and chemical analysis of the partially fused pellets. The compound should be subjected to dilatometry and differential thermal analysis in order to determine its properties more precisely.

Preparation of Low (α) Lithium Aluminate. The rhombohedral (α) form of LiAlO₂ is attractive as a target material because of its 30% higher density and the fact that it does not undergo hydrolysis as the tetragonal γ does.⁽¹⁾ However, obtaining pure α modification in large quantities is difficult.

Above 600 C the α form slowly and irreversibly transforms to the γ modification.⁽²⁾ Formation of the γ variety is also accelerated by nuclei of this kind.⁽¹⁾ Because of the difficulty of chemically forming the aluminate at such low temperatures, the pure γ or mixtures of α and γ are commonly produced by chemical vendors.

Attempts to chemically form the α variety have not been successful to date. A ball milled mixture of Li₂CO₃ and Al₂O₃ showed no reaction after 96 hours at 600 C. A similar mixture of LiOH and Al₂O₃ treated at 525 C for 20 hours produced a mixture of the α and γ forms as well as indications of incomplete reaction such as LiOH·H₂O and lithium spinel (LiAl₅O₈).

A further attempt will be made by heating an intimate mixture of LiOH and alumina in vacuum at 500 C. The vacuum will promote faster reaction and should exclude the reactants from water, carbon dioxide and other contaminants. Another possible reaction would be between lithium oxide (Li₂O) and alumina at temperatures substantially lower than even 500 C.

(1) Lejus and Collongues, Comp. Rend., 254, 2005-2007 (1962).

(2) Lehmann and Hesselbarth, Z., Anorg. Allg. Chemie, Band 313, 117-120 (1961).

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Capsule Irradiation of Lithium Aluminate and Lithium Silicate Targets.

Previous thermal extractions of tritium from irradiated lithium aluminate and lithium silicate failed to produce the calculated gas yields. To investigate the possibility of gas release during the irradiation, capsule experiments are being conducted. In the first experiment, pellets of the target materials were irradiated in quartz ampoules, with relatively large void volumes for gas accumulation. However, in postirradiation examination the ampoules were found to have broken. An aluminum ampoule in the same capsule had partially collapsed; the failures were attributed to overheating and concomitant gas release or to thermal expansion of the targets, causing the quartz ampoules to shatter.

A second capsule was irradiated for 4.4 days in the K-West snout facility. Estimated GVR's are 2.2 for aluminate targets and 3.0 for the silicate target. Targets are contained in three aluminum ampoules and in one quartz ampoule. The three aluminate targets included swaged, pressed and pneumatically-impacted samples; the silicate sample was prepared by swaging. A thermocouple inserted into a well at the center of the aluminum encapsulated silicate sample indicated 103 to 107 C during the irradiation.

In postirradiation examination the aluminum ampoules will be drilled to determine gas released during the irradiation. After drilling, one of the ampoules will be heated to 300 C to measure additional gas release. The targets will be retrieved to determine a complete gas balance by thermal extraction and chemical dissolution.

2. Corrosion and Water Quality Studies

Reactor Process Tube Hydriding. An experiment, designed to explore the current zirconium hydriding tendency in KE Reactor, was discharged and examined after 30 operating days at an average temperature of 104 C. Samples were precoated in various ways in order that judgment can be made of potential surface treatments as hydride barriers. The Zircaloy coupons were tightly held in a 6063 aluminum alloy holder. Metallographic examination of duplicate coupons showed excellent agreement between duplicate samples for form and amount of surface hydriding except for the anodized pretreatment where only one coupon of two hydrided. Hydrogen analyses on one set of coupons (30 to 40 mils thick) gave the following results:

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As-received and vapor blasted (process tube)	60 ppm
Etched	20 ppm
Etched and autoclaved (400 C, 1500 psi, 61 hrs)	None
Etched and anodized (100V 1% KOH)	27 ppm
Etched and air oxidized (500 C, 10 min)	44 ppm
As-received and vapor blasted (annealed sheet Zr-2)	615 ppm
As-received and vapor blasted (annealed Zr-4)	240 ppm

Out-of-reactor experiments utilizing 90 C process water and impressed potentials indicate that Zr-2 specimens charged at 3.0 and 4.5 volts (cathode) hydride rapidly while no hydride case has been observed on specimens held at 0.5 volts or 1.5 volts in 815 hours. Measurement of galvanic currents between aluminum and Zr-2 in reactor effluent is being planned. The effect of impressed AC potentials is also being studied.

Examination of K Reactors Zircaloy Process Tubes. Two Zircaloy process tubes have been examined for crud deposition and oxide thickness in conjunction with the investigation of the current hydriding problem in the K Reactors. One tube (2169 KE) had been in service since May 1963. The other tube (3075 KW) had been in service since August 1963. Rings were cut from the process tubes at various distances from the rear flange. Seven rings were cut from tube 2169 KE ranging from 18 inches to 43 feet. The seven rings cut from tube 3075 KW ranged from 2-1/2 feet to 30 feet from the rear flange.

The amount of deposited crud on each section of tube was determined by removing the crud with alkaline permanganate solution followed by ammonium citrate solution and measuring the resultant weight change of the section. The crud level on both tubes was found to be about equal. It was also found that the crud thickness increased considerably toward the rear of the tube, increasing from approximately 35 mg/dm² at 43 feet from the rear flange to approximately 195 mg/dm² at 8 feet from the rear flange. It was not possible to obtain crud thickness measurements on the final 8 feet of process tube due to crud losses in handling and tube discharge.

Oxide thickness measurements on the sections from process tube 2169 KE revealed extremely thin oxide film varying along the length of the tube. The oxide thickness at 18 inches to 8 feet from the rear flange was equivalent to 0.125 $\pm$ 0.05 mg/dm² weight gain. From 23 feet to 29 feet from the rear flange the equivalent oxide thickness weight gain was 0.25 $\pm$ 0.05 mg/dm². Beyond 35 feet, the equivalent weight gains were less than 0.100 mg/dm². The weight gains appear to reflect a radiation effect. The weight gains from the in-flux portion of the tube (0.25 mg/dm²) are

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a factor of two higher than the weight gains for the out-of-flux portion (0.125 mg/dm^2). The low weight gains of less than 0.100 mg/dm^2 for the tube section beyond 35 feet are probably due to low operating temperatures.

The oxide thicknesses were measured by an anodization technique in which the voltage required for current flow was established. This measured voltage was then converted to oxide weight gain by means of the conversion figure of $0.05 \text{ mg/dm}^2/\text{volt}$ for anodic films on Zr-2.

N-Reactor Fuel Element Deposits. The elements discharged from tube 2960 on June 6, 1965, were visually examined in the N-Reactor viewing basin. The first four elements in the upstream end of the fuel charge were covered with a uniform, thin layer of greyish black crud. The other elements in the charge appeared to have the normal shiny black ZrO_2 surface. These films are considerably different from the uniform reddish-brown film observed on fuel elements in previous discharges. The reasons for the varied films are presently unknown.

N-Reactor Decontamination Studies.

a. Decontamination Solution Variables Evaluation - The continuing study to improve reactor system decontamination processes is being directed toward evaluating the effects (on efficiency and corrosion) of variables in the alkaline permanganate + acid or salt procedure. The alkaline permanganate presently used is the "18-3" mixture (18% NaOH, 3% KMnO_4). A more dilute mixture would require less chemicals, be easier to use, simplify rinsing, result in less solid active waste to be disposed of, and probably be less corrosive. In preliminary beaker tests using sections from PRTR jumpers and from N-Reactor steam generator tubing, solutions containing 5, 10, and 18% caustic were equally effective in most cases. Additional tests are being scheduled to obtain results under more prototypical conditions.

The second step in the decontamination process employs an acid or a salt [at various times being H_3PO_4 , HNO_3 , NaHSO_4 , oxalic acid, sulfamic acid, and ammonium citrate at various pH values with or without EDTA]. Several of the reagents are ineffective or excessively corrosive. At present, three reagents are under test: Turco-4521 (a proprietary mixture of oxalic acid, ammonium citrate and other compounds), RD-16 (a proprietary compound of ammonium citrate + EDTA at pH 7), and Turco-4306-D (a proprietary compound containing principally sulfamic acid). The Turco-4521 is a significant improvement over other oxalic reagents (e.g., Turco-4518) previously used, primarily because it does not form an insoluble precipitate (except after 40 hours). In preliminary beaker tests, using portions of PRTR jumpers, the efficiency of the different procedures varied as follows: APOX (alkaline permanganate followed by Turco-4521) -

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300 to 600; APSUL (alkaline permanganate followed by Turco-4306-D) - 25 to 30; APACE-RD-16 (alkaline permanganate followed by RD-16) - 5 to 7. In these tests the APOX procedure was clearly superior; the APSUL gave much lower but still respectable D.F.'s; the RD-16 was not very efficient. In comparable tests using sections of N-Reactor steam generator tubing, decontamination factors were: APOX, 64; APSUL, 10; APACE-RD-16, 11.

The RD-16 is an attractive decontaminant because it is less corrosive, since it is neutral (pH ~7). However, the efficiency is not as good as for the Turco-4521, especially on the long-term films (e.g., PRTR jumpers). The efficiency can be increased by longer exposure; for example, for RD-16 after 6 hrs, the D.F. is 21. Longer exposure will, of course, also increase D.F.'s for other reagents; it will also increase corrosion.

b. Precipitation Effects - When a relatively complex system (such as a reactor) is being decontaminated, the decontamination occurs in two steps: (1) the adherent film is removed in the form of scale or sludge; and (2) the scale or sludge is dissolved. If the dissolution step does not occur, the undissolved scale or sludge will remain in the system. It may be dense or viscous and may be difficult to remove by flushing, especially with the extremely low flow rates generally available during decontamination. This scale or sludge carries much of the activity in it. Thus, a very low decontamination factor may result. It is therefore necessary that this dissolution step be obtained.

The time required for the dissolution step varies considerably with different decontaminants. In some cases, before dissolution is complete, a secondary reaction causing precipitation may occur. This effect was noted during examination of the surfaces of the N-Reactor steam generator after a recent decontamination. Considerable undissolved crud (essentially iron oxides) remained undissolved in the system. Evidently the decontamination factor could have been improved by using some solution which could dissolve and remove this crud. Three chemical decontaminants were tested to evaluate their capability for removing and dissolving the crud and film from portions of N-Reactor steam generator tubing. A pretreatment in 18-3 alkaline permanganate solution at 105 C for one hour was given all specimens. The samples were then rinsed and placed in a 50-ml graduate, which was then filled with the decontaminant solutions, thus giving a relatively large surface area for a very small amount of decontaminant. The tests are discussed in detail below:

Turco 4306-D: After treatment, undissolved flaky and flocculent material was observed settling to the bottom of the cylinder. The internal film on the tubing sample was only partially removed even

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though the color had been changed from black to red. About 95% of the activity was removed from the tubing, but some of the film remained as a sludge in the bottom of the graduate.

Turco 4521: After treatment the solution was a yellow color with no observed precipitate. All of the film was removed from the internal as well as external surfaces of the sample. About 98% of the film and activity were removed (D.F. ~ 60) and no undissolved sludge remained.

RD-16: After one hour the solution was speckled with flaky material. The sample was still filmed. After 2.25 hours, the sample was defilmed; undissolved material was observed settling to the bottom of the cylinder. About 98% of the film and activity were removed from the tubing, but a considerable fraction of the film and associated activity remained as sludge in the bottom of the graduate.

c. Film and Activity Studies - Analyses of the isotope distribution in the N-Reactor heat exchanger piping films before and after the decontamination were completed. The major isotopes found were Sb-124, Fe-59, Mn-54, Zr-Nb-95, Cr-51, Co-60, Ru-103, Ce-141, and Zn-65. The total amount of each isotope present was significantly reduced by the decontamination; however, some isotopes were more effectively removed than others. The Fe-59 composed 20 to 50% of the total activity after the decontamination, and Mn-54, Cr-51, Co-60, and Zr-Nb-95 each composed 10 to 20% of the activity.

Use of (PolyHall) Polyacrylamides to Reduce Corrosion and Friction. Laboratory tests were initiated to determine the corrosion inhibition characteristics of PolyHall compounds (polyacrylamides) which are supposed to reduce the friction loss in a flowing system. The tests were conducted at 95 C for 1400 hours. Three aluminum alloys were evaluated (1100, 6063, and X-8001) in three 20 ppm PolyHall solutions, P-27, P-159, P-295. The data indicate that P-27 is the most effective of the three compounds, reducing the corrosion rate by a factor of 2 to 3.

Hydride Formation in Zircaloy Tubes. Excessive localized hydriding has been observed at the rear sections of some Zircaloy tubes in the K-Reactors. The hydriding is concentrated on the inner surface of the rear three feet of the tubes examined; in most cases, a layer of zirconium hydride is formed. One possible mechanism includes both mechanical action and corrosion. The aluminum dummies in the rear sections, which are under heavy load, could wear away the protective oxide films by mechanical action (as by fretting), allowing unprotected zirconium and aluminum metal surfaces to be in intimate contact. The

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unprotected aluminum would corrode and the hydrogen formed during the reaction would be released at the surface of the unprotected zirconium. A test facility to test this hypothesis was operated for 540 hours. The samples discharged at that time showed considerable wear at the points of contact; samples are being analyzed for hydride. The test is scheduled to continue for another 14 weeks.

Scratched and aluminum-smeared samples of Zircaloy were exposed out-of-reactor in 6.6 pH process water at 120 C. Hydride cases 0.4 to 0.8 mil thick were found on samples exposed for 1200 hours. No hydride was detected on samples pretreated by etching, fluoride contact, or polishing alone, after 1200 hours of exposure.

The hydride found on these 1200-hour samples was similar in extent and appearance to that previously reported for 360-hour exposure. Since the hydriding did not increase with further exposure beyond the 360 hours, it appears that the presence of aluminum was an important factor in hydriding of these samples. The other factor which appears important to hydriding is mechanical damage of the Zircaloy oxide.

Hydriding is also present near the edges of Zircaloy samples exposed in aluminum spacers during other in-reactor corrosion tests; these samples were exposed in the 1706-KE single pass facilities for 89 days. Analysis of these and other in-reactor samples is still in progress.

CO₂ for pH Control: Testing of pH adjustment to 6.6 with 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 with pH reduced by CO₂ addition. Savings in chemical cost could accrue from use of CO₂ from stack gases to replace sulfuric acid for pH adjustment of reactor cooling water. Initial results from in-reactor corrosion samples indicate that use of CO₂ for pH adjustment provided good control of both aluminum and steel corrosion.

Half-Plant Test of Semi-Treated Water: Significant savings in costs of treatment chemicals, manpower, and equipment could accrue from reactor cooling with untreated Columbia River water, if radioactivity, corrosion, and filming are maintained at acceptably low levels. Large scale testing of raw water at F-Reactor included coupon corrosion samples installed in the rear sections of selected cooling channels. Treatment of this water included filtration, pH adjustment to 6.6, and inhibition with 1 ppm sodium dichromate, but did not include the coagulation and settling of fine solids. The test was terminated in May due to relatively high radioactivity in effluent water. Preliminary measurements from the in-reactor samples indicate that corrosion of aluminum and carbon steel is the same, or less, in this

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water, than in standard process water (at pH 6.6 with 1 ppm dichromate). Typical results for Aluminum Alloy X-8001 showed about 0.2 mils/mo in the control (process water) channels.

3. Gas-Atmosphere Studies

Oxidation Rate of N-Reactor Reflector Graphites. The thermal reactivity of TSX graphite (N-Reactor core material) with dry air at atmospheric pressure and in the temperature range from 625 to 727 C was reported in the April 1965 monthly report. The rate is given by:

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

Rates of the two reflector graphites (GLC and AGOT-LS) have now been determined under equivalent conditions and in the same apparatus. The rate for GLC can be expressed by:

$$R(\text{g/g/hr}) = (2.88 \pm 0.09) \times 10^{11} e^{\frac{-54.2 \pm 6.1}{RT}}$$

and for AGOT-LS:

$$R(\text{g/g/hr}) = (1.74 \pm 0.07) \times 10^{12} e^{\frac{-58.0 \pm 6.9}{RT}}$$

As would be anticipated, the less pure reflector graphites oxidized some two to three times faster than the core material.

Effect of Prior Oxidation on Reactivity. The reactivity of graphite increases with the extent of oxidation (percent burnoff) that a sample has achieved, at least for the first few percent. This increase has been attributed to an increasing surface area of the sample. Preliminary experiments here have indicated that the manner in which a given burnoff is achieved may affect the reactivity. To test this effect the following experimental program has been started.

Samples will be oxidized in increments of approximately 0.5% burnoff in CO₂, CO₂/CO mixtures, H₂O and H₂O/H₂ mixtures. Prior to the first oxidation and after each incremental oxidation the reactivity of the samples to approximately 15 mm of water vapor in the temperature range from 800 to 860 C will be determined. The results on samples with prior oxidation in 100% CO₂ at 860 C are presented below.

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EFFECT OF BURNOFF ON REACTIVITY TO WATER VAPOR

Burnoff in CO ₂ , %	Rate, hr ⁻¹		Apparent Activation Energy, kcal/mole
	800 C	850 C	
0.0 - 0.13	5.61x10 ⁻⁶	1.33x10 ⁻⁵	41.2
0.7 - 0.77	5.55x10 ⁻⁶	2.41x10 ⁻⁵	70.1

The large increase in apparent activation energy is somewhat surprising.

The Effect of Oxidation on the Sonic Modulus of TSX Graphite. Because the strength of TSX graphite has not been determined as a function of oxidation, the present study was initiated. While a definite correlation between the sonic modulus and either the mechanical modulus or strength has not been established, it is known that all three properties exhibit very similar trends as a function of oxidation. Since the measurement of the sonic modulus is nondestructive, it was selected as an indicator of strength, primarily to avoid differences between graphite samples, i.e., a given sample can be employed for an entire series of measurements.

To date, measurements have been completed on two parallel samples of TSX graphite. Oxidation is performed in air flowing at about 1 ft³/hr. Different oxidation temperatures are employed for each sample to determine the effect of the rate of oxidation on the modulus. The results are compared in the following table to data reported by Rounthwaite et al for PGA graphite oxidized in CO₂.

Sample No.	Oxidation Temp., °C	Relative Modulus, E/E ₀	Oxidation Range, %
213-231	595	0.966 ($\frac{w}{w_0}$) ^{7.20}	0 to 20.7
213-232	646	0.980 ($\frac{w}{w_0}$) ^{8.17}	0 to 18.8
PGA	770	0.993 ($\frac{w}{w_0}$) ^{8.60}	0 to 15

Column 3 gives the relative modulus, E/E₀, as a function of the fractional weight of sample remaining. It is seen from the slope of these logarithmic equations that TSX graphite compares favorably with PGA, the loss in modulus with oxidation occurring less rapidly in TSX. Also, the modulus appears to decrease more rapidly for the sample oxidized at the higher temperature; this could result from sample differences.

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Calculated values of the utilization factors indicate that oxidation is essentially homogeneous in both samples.

4. Process Tube Development

Fracture Studies. A facility for studying the bursting characteristics of pressure tubing at subzero temperatures has been fabricated. It consists of an insulated polyethylene vessel in which a methanol bath can be cooled with dry ice. Temperatures slightly below -100 F are attainable and are measured by reversing the leads on an ordinary iron-constantan thermocouple affixed to the test specimen. The tube undergoing test is pressurized with a glycol mixture that will resist freezing at the test temperature. The immediate area of the low temperature bath is blanketed with CO₂ as a precaution against ignition of the methanol by a chance spark that might be generated by the bursting specimen.

A stress-strain curve has been plotted from strain gage readings taken on a specimen of N-Reactor pressure tube pressurized to failure at -33 C. The 0.2% yield strength and the ultimate strength were virtually coincident at 115,000 psi. This compares with 105,000 psi at room temperature, 80,000 at 150 C, and 60,000 at 300 C. These four values form a nearly linear relationship between yield strength and temperature within this range. Extension of the yield strength curve to the subzero region is expected to be helpful in forthcoming efforts to determine the nil ductility transition temperatures of the various types of pressure tubing.

5. Thermal Hydraulics Studies

Hydraulic Tests for the Present Production Reactors. Experiments were performed to determine the pressure drop behavior of two, 1-inch OD pigtails used in conjunction with a K-Reactor front nozzle adapted for use on the rear of B-D Reactors. One of the pigtails contained an extra loop to reduce stresses resulting from the thermal expansion of the process tube. These tests revealed little difference in pressure drop between this pigtail combination and the standard pigtail.

In other experiments an evaluation was performed of the ability of Jaguar MD-A (a natural polymer) to reduce frictional losses in a piping system. Studies performed by the Navy have indicated that adding as little as 1/2% of similar substances to water flowing in a pipe could reduce the usual turbulent friction head loss by as much as 70%. The application of Jaguar MD-A, Separan, or some other similar substance is being considered as a possible means of increasing the capacity of the reactor coolant intertie system.

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A sample of Jaguar MD-A was tested in a closed loop system to determine the effects of various concentrations on the pressure drop through 1/2-inch copper tubing and 1/4-inch iron pipe. It was found that concentrations of 125 ppm resulted in pressure drop reductions of 20% in turbulently flowing water, and concentrations of 500 ppm gave reductions of 50 to 60%. The pressure drop was observed to slowly increase with time which indicated that this particular material may not be satisfactory for continuous use in a recirculating loop. On a "once-through" basis, however, this material could significantly increase flows in a piping system. Before such substances can be used in the intertie system, further studies are necessary to determine mixing, storage, and inspection methods and to determine the effects of such substances on convective heat transfer.

6. Graphite Studies

N-Reactor Graphite Irradiation. The two final capsules, H-5-5 and H-6-5, in the N-Reactor graphite irradiation program were removed from the GETR during the week of June 14. Maximum neutron exposure is estimated to be 9×10^{21} nvt, $E > 0.18$ MeV. Studies indicate it is not feasible from a technical or cost standpoint to continue these irradiations in the ETR or at Savannah River and consequently this program is now terminated.

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 High Power Density Core. The HPD fuel testing program in support of the planned Molten Core Experiment is proceeding on schedule. The maximum burnup on HPD fuel elements currently in the PRTR is 4190 Mwd/ton. This maximum burnup was accumulated in the core of the reactor at maximum rod power generation rates of approximately 19 kw/ft (maximum fuel temperature ~ 2600 C).

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Special HPD test elements are being irradiated at higher powers in the FERTF. The UO_2 -2 wt% PuO_2 HPD thermocoupled fuel element (FE-6502) operated successfully in the FERTF at a tube power of 1525 kw for six days. This tube power produced an estimated maximum rod power generation rate of 21.5 kw/ft with fuel temperatures above melting during steady state operation. Postirradiation examination of three nonthermocoupled rods from this element is in progress. Diameter measurements indicate that no discernible changes occurred as a result of irradiation. One hundred and eight ml of gas (STP) were collected from each of two rods from the 12-rod ring. The gas collected (somewhat less than the amount expected to be released) would produce a calculated maximum internal pressure of 422 psi. Examination of the postirradiated fuel structures and autoradiographs indicate that fuel was molten to approximately 13% of the fuel radius, to agree very well with predictions. This indicates that in-reactor fuel sintering rates are sufficiently rapid to avoid higher-than-normal fuel temperatures during startup.

Other pertinent phenomena observed during the examination of these rods are as follows:

1. The once-molten fuel contains a high concentration of fission products.
2. There is evidence of liquid fuel having been extruded into radial cracks that existed during operation. Other cracks show no evidence of once-molten fuel.
3. In addition to the normal fission product migration patterns, a high concentration of plutonium-bearing particles appears to have formed at the extreme outer sintering boundary.
4. There is no evidence of axial fuel relocation.
5. The inclusion of graded enrichment markedly reduced the fuel temperatures in the end cap regions.

A preirradiated, vibrationally-compacted HPD element defected with a 1/16-inch diameter hole operated successfully for approximately 80 hours at a maximum tube power of 1425 kw with incipient melting (20 kw/ft) at the point of defect. Prior irradiation of this element (19 kw/ft maximum) was sufficient to cause considerable in-reactor sintering. Burst type activity releases, characteristic of a leaker defect, occurred during incremental power increases at the lower power levels but not at the higher levels. In all cases, during continued operation, the loop activity approached a steady value in an asymmetric

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manner. The steady state GM activity in the loop during the irradiation of this element was approximately 40 times greater than for any defected element previously tested. The rod power, however, was three times greater than for any previous experiment. Activity bursts when the reactor was shut down and when the loop was depressurized were observed. No changes in the physical appearance of the defect were observed during postirradiation examination in the basin. A grey discoloration of the cladding downstream from the defect, due to the evolution of material and/or gases from the hole, was apparent. Continued irradiation of this element at higher rod powers is planned.

Short-core Fuel Element Cladding. Bids were received on the additional 1400 tubes ordered for fabrication of short-core fuel rods. Because of exceptions taken by the vendors, the requisition was re-phrased and submitted for rebids. Delivery of these fuel tubes is now expected in mid-August.

PRTR Fuel Element Performance. The present PRTR fuel loading consists of the following elements:

<u>No.</u>		<u>Enrichment</u>
8	Includes a salt cycle element and the 2 wt% PuO ₂ defect element.	UO ₂ -2.0% PuO ₂
1	EBWR test element.	UO ₂ -1.5% PuO ₂
35		UO ₂ -1.8% PuO ₂
1		UO ₂ -0.75% PuO ₂
31		UO ₂ -0.48% PuO ₂
3		UO ₂ (natural)

The maximum fuel exposures as of July 1, 1965, are as follows:

		<u>Fissions/cc x 10²⁰</u>	<u>Mwd/ton_{fuel}</u>
UO ₂ -0.48% PuO ₂ (Incrementally Loaded)			
Vibrational Compacted	FE-5118	2.46	8910
Swaged	FE-5142	2.30	7500
UO ₂ (natural)			
Vibrationally compacted	FE-1097	1.56	6077
(retired)			
Swaged	FE-1037	2.17	7850
UO ₂ - 0.75% PuO ₂ (Incrementally Loaded)			
Swaged	FE-5221	1.30	4250

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		<u>Fissions/cc x 10²⁰</u>	<u>Mwd/tonfuel</u>
UO ₂ -1.0% PuO ₂ (Pneumatically Impacted)			
Vibrationally Compacted	FE-5227	1.32	4650
Swaged	FE-5224	1.87	6100
UO ₂ -1.5% PuO ₂ (Pneumatically Impacted)			
Vibrationally Compacted	FE-6501	0.53	1850
UO ₂ -2.0% PuO ₂ (Pneumatically Impacted)			
Vibrationally Compacted	FE-6001	1.03	3640
Swaged	FE-6002	0.82	2770
UO ₂ -2% PuO (Salt Cycle Fuel)	FE-6500	0.25	885

One fuel element failed during reactor operation. The incrementally-loaded, swaged, UO₂-0.48% PuO₂ element FE-5192 failed on or about 5/16/65. The element continued to operate in the reactor for approximately eight days without hampering reactor operation. When the reactor was shut down, the element was discharged and examined in the basin. Gas bubbles were seen issuing from a discolored (ZrH_x) area in the Zr-2 cladding, 18 inches from the top of the rod. The rod was removed and sent to Radiometallurgy for destructive examination. The fuel exposure is 1.69×10^{20} fissions/cc (5550 Mwd/tonfuel).

UO₂-Water Reaction Studies. Molten UO₂-water reactions are being investigated as part of the over-all safeguards analysis for the Molten Core Experiment in the PRTR.

Current bench-top experiments are designed to simulate reactor conditions of water temperature and pressure. Uranium dioxide bars are melted by self-resistance heating and subsequently dropped into heated static heavy water. The system is contained in a high pressure furnace and maintained at a variable argon pressure during the reaction. The parameters being investigated are: (1) maximum pressure increase, (2) rate of pressure rise, (3) rate of water temperature increase, (4) UO₂ particle size distribution following reaction, and (5) extent of UO₂ oxidation as revealed by deuterium release. The variables will consist of: (1) UO₂ temperature prior to contacting the water, (2) argon pressure, and (3) water temperature.

Fabrication Cost of Plutonium-enriched Fuel Elements. The variable cost of fabricating plutonium-enriched fuel rods for the Experimental Boiling Water Reactor (EBWR) is \$198 per kilogram of fuel (as oxide) or a unit cost of \$164 per fuel rod. The shop labor requirements for

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each rod is 5.53 man-hours direct labor and 1.61 man-hours indirect labor. The variable costs include direct materials, some processing costs for nuclear materials, shop labor, direct overhead, and consumable supplies but do not include the cost of nuclear materials themselves, use charges, administrative overhead, research, development or engineering costs.

Each fuel rod contains 830 grams of depleted UO_2 -1.5 wt% PuO_2 fuel. The core length is 123 cm. The rods are 1.0 cm OD with 0.06 cm Zircaloy cladding. The costs are based on the fabrication of the last 200 rods by a well-developed, pneumatic impaction-vibrational compaction process. Thus, the costs represent the current "state of the art" and do not reflect the historic cost of the entire fabrication.

High Exposure Plutonium Studies. A study was initiated this month, in cooperation with Chemical Separation personnel, to obtain information that will be useful in handling and fabricating elements containing high exposure Pu.

There are 16 fuel element assemblies from the PWR Shippingport, containing high exposure Pu, at Hanford for separation. Additional assemblies are due by July 1, 1965.

Irradiation history is now being compiled on these assemblies. Selected fuel rods will then be removed from the assembly and the Pu recovered. Isotopic analysis and microshielding studies will then be made on the Pu.

An attempt was made to separate one of the assemblies (7 bundles of 120 rods each), but the special tooling used in the underwater handling will have to be modified. Another attempt will be made before July 1, 1965.

The Radiometallurgy Laboratory will be ready to receive the bundles as they are separated from the assemblies.

FD-15, Safety Rods. As part of the Physics Fuel Fabrication Program, 12 safety rods for the PRCF were fabricated this month. These safety rods are for the third light water core of the PRCF. The safety rods consist of 49-inch long, 0.030-inch thick cadmium tubes with Plexiglass cores placed in Zircaloy tubes. The poison rods were attached to regular PRCF fuel rods containing 2 wt% PuO_2 - UO_2 , wherein the PuO_2 has an isotopic content of 8% Pu-240. Special end caps were fabricated as described in SK-3-11566 to facilitate handling in the critical facility.

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Fuels Status. Since January 1965, several programs have required the use of extensive quantities of mixed oxide compacted $\text{UO}_2\text{-PuO}_2$ and some pure PuO_2 . Some of these special blends have required extensive development of tooling and processing techniques. A summary of these programs and the enrichment levels are contained in the following table:

Pneumatic Impaction of UO_2 and PuO_2
January 1, 1965 to July 1, 1965

<u>Program</u>	<u>$\text{UO}_2\text{-PuO}_2$ Grams</u>	<u>PuO_2 Grams</u>
Saxton @ 6.6% PuO_2	120,000	7,920
FRTR @ 2%	1,266,000	25,320
@ 4%	120,000	4,800
EBWR @ 1.5%	66,000	990
PCTR @ 2.7%	89,000	2,403
PRCF @ 2.0%	1,571,000	21,420
ANL @ 100%	--	2,674
Misc.	<u>19,520</u>	<u>3,855</u>
Total	3,355,500	69,382

Diffusion Test Element. The second diffusion test element is under irradiation in the MTR B-3 loop. This test will operate at higher surface and central temperatures than the first diffusion test. The element, GEH-4-117, will be discharged in July and returned for examination.

In-Reactor Fission Gas Pressure Measurement. Scope design of in-reactor fission gas pressure measurement apparatus for Pu-enriched fuels was completed. The design will incorporate the null-pressure balance type pressure transducer successfully used at GE-Vallecitos Atomic Laboratory on UO_2 fuel elements. Detail design has begun.

Defect Propagation in Zr-4 Cladding. Evaluation continued of the effect of thermal cycling on fabricated fuel rods containing defects in the cladding. Three fuel rods fabricated from rejected Saxton tubing were subjected to 45 thermal cycles. Metallurgical examination is scheduled after 50 thermal cycles.

Chloride-Zircaloy Cladding Compatibility. Four MTR test capsules, fabricated to determine the effects of chloride on Zircaloy-2 cladding during irradiation, are scheduled for irradiation in the MTR. The capsules contain 100, 200, and 300 ppm chloride concentrations,

respectively, in a UO_2 -2 wt% PuO_2 fuel. The fourth rod, containing less than 10 ppm chloride, is used as a control.

Recent irradiations of salt cycle fuel containing from 50 to 90 ppm chloride impurities indicate that the present fuel specification limit of 10 ppm chloride may be much lower than necessary.

The capsules were sent to the Idaho Falls Testing Station and are scheduled for irradiation in the next MTR cycle. They will be irradiated to 3000 to 5000 Mwd/ton. One capsule may be replaced after one cycle in the MTR and returned to Battelle-Northwest for post-irradiation examinations.

Spheroidization Studies. Continuing studies on the spheroidization of UO_2 was concentrated on a method that provides a large quantity of particles with a minimum of fabrication steps. Carbowax binder was added to a five-pound batch of ceramic grade UO_2 , using a Hobart mixer. The UO_2 was then blended in a twinshell blender while still wet. The complete batch formed into agglomerated particles.

Blending time controls the size and the variation of sizes within the batch. The longer the blending period, the more uniform the particle size. A blending time of about 30 minutes gives a batch of 75% -6 +10 mesh particles. A shorter blending time results in a size variation such as is shown in the following table.

Screen Analysis for UO_2 Spheres

<u>Mesh Size</u>	<u>% Retained</u>
+6	23.35%
-6 +10	21.73
-10 +20	28.52
-20 +35	18.57
-35 +65	6.45
-100	1.17

The spheres were sintered in flowing hydrogen at 1750 C for 12 hours. The first batch had a low density (72% at 9); however, it is anticipated that different sintering and blending techniques should result in higher densities.

In other work, mixtures of UO_2 powders with paraffin or stearate binders were pressed through a perforated steel plate (1/7-inch diameter holes) and cut into 1/8-inch long chunks. These chunks were spheroidized in rotating alumina jar mills using the following

spheroidizing surfaces:

1. Alumina jar.
2. 320-C grit wire screen liner.
3. 320-C grit emery cloth liner.

All three surfaces rounded the green chunks, but the wire screen treatment resulted in rough surfaces on the UO_2 particles. Mercury porosimetric tests on -6 +45 mesh particles indicated densities (1.9 micron pore size) of 92.6 to 94% of theoretical density.

Remote Equipment Construction - Fuels Recycle Pilot Plant. The 3-D model of the 324 Building Metallurgical Facility is essentially complete and is currently being used as a tour and equipment design aid.

Powder preparation equipment consisting of gyratory crusher, magnetic separator, screens, vibrational feeders and pneumatic compactors were received on site. Work on unitizing this equipment for remote hot cell operation began.

New decladding equipment for the hot cell facility was received. Decladding equipment will be provided for maximum removal of fuel material from the cladding and also for minimum damage to the cladding.

Tooling. Tooling for underwater changing of EBWR fuel rods was developed. There will be a rod removed from a special EBWR element that has been irradiated in the PRTR. A newly fabricated cold rod will be installed in the element. This work will be scheduled in the PRTR basin in the near future.

Corrosion and Water Quality Studies

Corrosion of Aluminum in Dilute Boric Acid Solutions. An in-reactor probe to study the effect of dilute boric acid solutions on the corrosion of aluminum alloys and the deposition of boron on the aluminum surfaces has been designed and constructed. It will be installed in PRTR in the near future (PRTR Test 122).

PRTR Boron Shim Studies.

a. H_3BO_3 Prototype Installation - The PRTR Design Test describing and authorizing the Phase 1 tests of the H_3BO_3 prototype PRTR was approved, and two of the four ion exchange units for this test have been deuterized. The prototype will be connected to the moderator piping system about the end of June, and the first Phase 1 test is scheduled to start in the next two or three weeks. Schedule for the second test is contingent on the progress of the outage and the results of the first test,

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but the test is tentatively scheduled for mid-July when the reactor resumes operation.

The automatic boron analysis unit operates satisfactorily except for a minor sampling problem, which is being corrected. Calibration of the analyzer system will be performed with D₂O solutions early next week.

b. H₃BO₃ Ion Exchange Studies - A method for recovering H₃BO₃ from exhausted ion exchange resins is required to reclaim the B-10 from the resin and thus make the use of B-10 enriched H₃BO₃ economically feasible at PRTR. As reported previously, H₂SO₄ and Na₂SO₄ solutions are effective regenerants for H₃BO₃; however, separation of the resulting borate-sulfate solution is quite involved. NH₄OH is not efficient enough to effect quantitative boron recovery with a reasonable volume of regenerant. Initial tests with 10 wt% NaOH indicate both quantitative recovery and high efficiency. Purification of the resultant solution by simple cation exchange appears feasible. Work is continuing to evaluate this process.

Tests to determine anion resin boron capacity values under dynamic conditions were continued to obtain more quantitative information. It appears now that the design previously recommended for the ion exchange system in the PRTR H₃BO₃ prototype unit is quite conservative. This conclusion was confirmed by laboratory model tests simulating full-scale prototype tests.

c. Corrosion Effects in Borated Water - The PRTR dump valve test in a helium atmosphere over a 50 ppm boron solution at 140 F has continued for a total of two months of periodically wetting and actuating the springs. Operation continues to be normal. The dump valve will be dismantled and examined during the next reporting period.

The shim well in the EDU Shim Test Loop was replaced and the pilot plant corrosion test continued at 50 ppm boron solution and a temperature of 140 F with no flow to the bottom of the shim well. The Inconel shim rod was forced into contact with the 6061 aluminum shim well. An examination of the shim well after one month of testing revealed only minor scratches made when charging the shim rod.

Inspection of corrosion coupons after a three-month exposure to 50 ppm boron solution at 140 F revealed pitting between galvanic coupled 6061 aluminum and Inconel. The 6061 aluminum had two pits approximately 60 mils in diameter and 30 mils deep while the Inconel penetration was negligible. The carbon steel coupons discharged to date indicate small localized pitting and a maximum penetration of two mils when

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calculated as uniform corrosion. The remaining aluminum and stainless steel coupons indicate negligible penetrations.

Continuous Corrosion Monitoring: Corrosion is presently being measured continuously with corrosion probes in three locations in PRTR systems. An aluminum probe element at a reflector coolant pump continues to show a corrosion rate of about 0.02 mil/month (this probe was installed on April 22, 1961). A probe element of 6061 aluminum at a moderator pump presently shows a corrosion rate of about 0.1 mil/month, while a carbon steel probe element in the bottom shield outlet shows a rate of 0.03 mil/month in this pH 10, 60 C water. These latter two probes were installed on April 29, 1965.

Decontamination of PRTR Jumpers: Jumpers are removed periodically (one every 13 months) to evaluate activity buildup and decontaminability. Differences in decontaminability of these jumpers are striking. This can best be illustrated by examples. Jumper 2049 was exposed for 26 months (Nov., 1962 to Jan., 1965) and was decontaminated on June 9, 1965, using the APCX procedure. The activity level before decontamination was 600 mrad/hr; the decontamination procedure removed activity down to background. Jumper 12-9 was exposed for 22 months (Nov., 1962 to Sept., 1964) and was decontaminated on June 9, 1965, using the identical procedure for Jumper 2049. The activity level before decontamination was 150 mrad/hr, and the activity level after decontamination was 28 mrad/hr. Thus, for Jumper 2049, the DF was >600, while for Jumper 12-9, the DF was 5.4. The results were checked using nine different decontamination procedures and results were all consistent (600/5.4, 385/5.0, 270/6.1, 7.0/1.9, 5.0/2.0, 5.7/1.9, 28/2.9, 23/3.9, and 27/3.5). It appears that there is an essential difference in the characteristics of the activities on the two jumpers. One plausible hypothesis is that some off-standard condition occurred during exposure of Jumper 2049 that resulted in deposition of a less adherent film. This hypothesis is being checked. It is interesting to note that both jumpers had been installed prior to the 1962 decontamination.

PRTR Pressure Tubes

Process Tube Studies: Work in progress on PRTR tubes consists of stress-rupture studies, crack propagation studies, measurement of distribution of elastic strain on a tube specimen with closed ends, measurement of elastic constants of tube materials, and determination of the time-temperature influence upon irradiated tube material.

One thousand hours at 550 F have reduced the ultimate strength of irradiated PRTR tube specimens by about 4% compared with specimens which had been heat treated 40 hours at 550 F temperature.

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Elastic strain measurements on process tubes indicate no significant difference between the elastic strain at two inches distance from the end fitting compared with 11 inches. The practical consequence is that PRTR tube burst test specimens of significantly shorter lengths may be used with confidence.

Reactor Components Development

Fretting Corrosion Investigation. Inlet jumper vibration monitoring records from the PRTR, during the report period, have indicated several characteristics not previously observed. In addition, the amplitude of vibration has, on occasion, been greater than 10 mils average peak-to-peak. The table which follows presents average peak-to-peak amplitudes during various modes of operation.

<u>Date</u>	<u>Reactor Power, MW</u>	<u>Avg. Peak-to-Peak Amplitude, mils</u>		
		<u>Tube 1354</u>	<u>Tube 1249</u>	<u>Tube 1653</u>
5/11/65 (1600 hrs)	70	6.55	8.45	9.0
5/16/65 (1800 hrs)	70	10.0	9.3	8.9
5/31/65				
(a) Scram - 127 min	30	7.72	8.6(1)	4.4
(b) Scram - 52 min		7.72	5.4(2)	4.5
(c) Scram - 42 min		7.70	7.6(2)	4.6
(d) Scram - 20 min		7.10	10.0	4.65
(e) Scram - 1 min	35	7.32(3)	10.0(4)	4.55(4)

- (1) Inlet jumper motion of tube 1249 started to increase at approximately 1.3 mils per hour.
- (2) Reported values are taken from the "valley" of an instantaneous decrease. The vibration level then immediately increased to the amplitude observed prior to the decrease.
- (3) Amplitude increased to approximately 8.9 mils immediately following the scram.
- (4) Amplitude decreased immediately following the scram.

The referenced scram was caused by a primary coolant system flow transient.

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Bearing failures in the EDEL-I circulation pump assembly have delayed completion of the 500-hour high flow test on the PRTR full length fuel element assembly. Loop flow conditions have been 182 gpm, 530 F, and 1050 psig for 186 hours. Pressure tube inspection after 93 hours of operation did not indicate any fretting attack. Repairs have been completed and operation resumed.

Shroud Tube Removal. Detailed design of the main machine for PRTR calandria shroud tube removal is 90% complete, and fabrication of the machine has started. Fabrication of the cutting heads is in progress. Scoping has started on an auxiliary device to remove portions of the bearing blocks between the calandria and bottom shield. This latter device will be used to locate the source of light water leakage from the bottom primary shield.

D2O Gasket Seals. The gasket test program seeking improvement in the sealing qualities and in the longevity of the spiral-wound, asbestos-filled, D2O gasket seals on the PRTR pressure tube assemblies has continued. Since stress corrosion cracking is thought to be the primary cause of gasket leakage and short gasket life, it is one of the major points of investigation. Two sets of stainless steel wound gaskets were given a laboratory treatment to produce incipient stress corrosion; an accompanying set of Inconel wound gaskets exhibited no stress corrosion cracking. All three sets were installed in the EDEL-I gasket test station and given 20 simulated PRTR thermal cycles in 83 hours. There was essentially no leakage for either type of gasket at the inlet and nozzle-to-tube locations. There was a small amount of leakage (avg. 0.5-1.4 ml/hr) at the cap gasket for the two stainless wound and the one Inconel wound gasket. The gaskets are being removed for examination and another group of three sets including stainless steel gaskets moderately stress-corrosion cracked are being prepared for testing.

In the two tests thus far, each involving 20 simulated PRTR thermal cycles, the Inconel wound gaskets have provided at least as good sealing at the inlet and tube-to-nozzle locations as the stainless steel wound gaskets. Although the leakages at the nozzle cap have been small (~ 3.5 ml/hr), the three Inconel wound gaskets have leaked more than the stainless steel wound gaskets.

Second Generation Mechanical Shim. The order for three new zirconium lead screws has been modified to incorporate wipers on the top of each ball nut. These wiper inserts will be a split-ring type, slightly spring loaded against the screw; the purpose being to eliminate particulate fouling within the ball nut, thought to be a major cause of shim rod sticking in PRTR.

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Design Analysis

Gamma Heating in PRTR Shield Plugs. During conversion of the PRTR to high power density, 13 outer channels will not be fueled. Current plans are to remove the process tubes from these channels and install shield plugs in the top and bottom primary shields. Gamma heating of these shield plugs is being evaluated to determine if cooling is required for the plugs. Calculations to determine gamma flux at the shield plug from capture and core gamma radiation at the shield plug have been completed. Streaming calculations were done for the empty, 4-1/4" diameter process channel using the methods reported in document APEX-525. The streaming flux reaching the top of the shield plug has been determined to be about 49% of the total flux at that location. Heat deposition rates and heat transfer calculations are now under way.

PRTR Chemical Shim. At the request of Nuclear Safeguards and Engineering Section, a calculation was made of potential transients in reactivity and boron concentration resulting from a postulated rupture of a cold, pressurized pressure tube in the PRTR with the moderator containing 140 ppm (weight) natural boron. The calculations were based on a MIDAS computer program solution for arbitrarily normalized boron concentrations previously applied to lower postulated boron concentrations. For the 140 ppm case, with the moderator level at 108 inches, initial changes in concentration and reactivity were calculated to be 1.2 ppm/sec and 1.4 mk/sec, respectively; at a 72-inch moderator level these rates would be 1.7 ppm/sec and 2.1 mk/sec, respectively. At the end of 40 seconds, boron concentration would decrease to 112-114 ppm for the 108-inch moderator level, and 107-110 ppm for the 72-inch level. The corresponding reactivity contributions from boron dilution in these cases would be about 38 mk at a 108-inch moderator level and 47-50 mk for a 72-inch level.

The calculations indicate that the reactivity ramp resulting from a boron dilution accident at a 140 ppm initial natural boron concentration would not be much greater than those previously calculated for lower concentrations (1.3-1.9 mk/sec at 86.4 ppm initial concentration), and should not appreciably alter safeguards requirements from the previous case.

PRTR High Power Density Core Kinetics and Control. Calculations of neutron kinetic parameters and mechanical shim rod strength have been completed for a revised set of nuclear data for the HPDC. The revised data incorporate 140 ppm natural boron concentration as a maximum in the moderator instead of the 86 ppm previously considered. The net effect of the increased poison was to increase the strength of the centrally located shim rods slightly (~ 1 mk/rod) and to decrease the

outer shim strength such that the total rod system strength is essentially unchanged from that calculated for 86 ppm. The prompt lifetime is expected to decrease slightly, and the delayed neutron fraction will remain the same.

Plutonium Utilization. Calculations are continuing on the nuclear characteristics obtained in a power reactor modeled after Yankee Core I in which plutonium is substituted directly for uranium-235 enrichment in the fuel. In addition to the case reported last month where low exposure plutonium (5% Pu-240) is utilized, a second case using high exposure plutonium (20% Pu-240) was investigated. Assuming the same fuel enrichment (0.4% U-235 + 3.0% Pu), the initial reactivity was reduced nearly 15% Δk , leaving insufficient excess to sustain significant exposure.

Thermal Hydraulic Studies

Consideration was given to the hazards associated with the failure of a jacket on a PRTR fuel element operating at a power density where molten fuel could occur in the interior of the element. At high burnups, sufficient fission product gas pressure could be present within the fuel to cause a rapid injection of molten UO_2 into the coolant if a cladding failure should occur. When this molten UO_2 contacts the coolant, large quantities of steam would be rapidly formed and conceivably could result in the rupture of the pressure tube or other coolant system components as a result of the pressure transient.

A detailed analysis is being performed to define the magnitude of the pressure transient associated with this potential release of molten UO_2 to the coolant. Preliminary calculations have shown that the transfer of heat from the UO_2 to the water would occur with sufficient rapidity to cause most of the steam to be formed in the pressure tube. As a result, very high transient pressures could occur there. More detailed calculations are being performed to refine the analysis further.

2. Plutonium Ceramic Research

Differential Thermal Analysis. During differential thermal analysis of PuO_{2-x} , a small, distinct peak was observed at approximately 875 C with specimens having O/Pu=1.55. It is proposed that this is a result of the $\alpha - \alpha'$ reaction which had been postulated to occur below 900 C. The reaction is exothermic on heating. By comparison of peak height with that for the $\alpha - \beta$ quartz inversion, the heat of reaction appears to be quite small, in the range 10-30 cal/mole.

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The problem of specimen oxidation has been brought under control, and a new series of specimens is being prepared for analyses.

Phase Studies. Measurement of oxygen dissociation pressures over americium suboxides was continued. Two additional isotherms at 1190 and 1400 K were determined. At the former temperature five days was required to reach equilibrium at a composition of $\text{AmO}_{1.915}$. Data from the previous isotherms were reproduced to within 0.003 O/Am units. The partial molal enthalpy of solution of oxygen was found to vary with composition, ranging from -120 kcal/mole at $\text{AmO}_{1.98}$ to -96 kcal/mole at $\text{AmO}_{1.86}$.

Additional plutonium suboxide powders of several compositions were prepared for thermal analyses and high temperature x-ray diffraction studies. Calibration of the 19 cm Unicam camera was begun. Several high density suboxides also were prepared by reducing impacted PuO_2 in hydrogen. Ceramographic examination of these samples revealed a small grain size with a large number of cracks in certain areas, as well as density variations; i.e., large regions of high and low density resulting from the Dynapak process.

Pneumatic Impaction of PuN. A procedure was devised for loading a capsule containing a small quantity (50 gm) of PuN into a standard Nu-pac can while maintaining the material in an inert atmosphere. Impaction at 1200 C yielded a PuN density of 93% of theoretical. Several pieces were recovered with as much as 7 mm across a face. X-ray diffraction analysis of the impacted material indicated a slight increase in lattice parameter (0.003A). The PuO_2 peak, barely discernible in the starting material, was even less evident in the impacted material. A solution reaction might have been responsible for these changes.

Preliminary ceramographic examination of the dense material revealed a few isolated inclusions tentatively identified as free Pu metal. Small pores appear to have concentrated at grain boundaries, leaving most larger grains free of porosity. No distinct oxide phase was detected.

High Density PuO_2 Research Specimens. Thin discs of several thicknesses were prepared from pneumatically impacted PuO_2 , approximately 98% of theoretical density, for basic studies such as thermal diffusivity measurement. The finished specimens are approximately 0.250-inch diameter by 0.030-inch, 0.040-inch, and 0.050-inch thick. Another specimen is being prepared from a large PuO_2 boule for thermal expansion measurements.

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Thermionic Work Function Measurements. A computer program was written in FORTRAN IV to analyze thermionic emission data and to provide more accurate calculation of work function and emission constant. This program is being debugged and should be ready for use in a month.

A new emission apparatus was assembled to replace a collector that had become contaminated because of a small leak in the vacuum system. A new emitter also was installed to provide improved mechanical stability. The new system is now being baked out.

PuO₂-Stainless Steel Cermet Irradiations. An eight-pin cluster of cermet pins successfully completed its scheduled irradiation in the ETR P-7 Loop. The pins operated at a calculated maximum power of 1.14 kw/in. (maximum core temperature of 850-1000 C) and achieved a maximum burnup of about 4.5% of the heavy atoms. The cluster included low density, cold pressed cermets (80% TD) and high density, extruded cermets (95-97% TD). Core compositions included SS-20 vol% PuO₂, SS-20 vol% beta-Pu₂O₃, and SS-30 vol% UO₂. The cluster will be returned to Battelle-Northwest for post-irradiation examination.

Irradiations of high and low density cermets are continuing in the MTR. Specimens in the reactor have reached a burnup of about 4.5% of the Pu atoms and are scheduled to reach a burnup of 6%.

PuN-316 Stainless Steel Compatibility. Irradiation test capsules are being fabricated to investigate the compatibility of PuN and type 316 stainless steel during irradiation. Eight PuN-20 wt% type 316 stainless steel pellets were pressed and loaded into stainless steel cans for densification by pneumatic impaction. The eight fuel capsules will be impacted in a specially designed die to provide appropriate dimensional control.

Oxidation problems experienced in the past were circumvented by minimizing the exposure of PuN to the nominally inert hood atmosphere. An x-ray diffraction analysis showed no evidence of PuO₂ contamination of the fuel pellets.

3. Ceramic (Uranium) Fuel Research

Thermal Diffusivity of UO₂. Thermal conductivity of polycrystalline UO₂ calculated from thermal diffusivity and reported heat capacity data show excellent agreement with previously reported thermal conductivity data. The pulsed energy technique was used to measure thermal diffusivity of the same polycrystalline UO₂ specimen on which the thermal conductivity was measured by comparative flow techniques at Battelle-Columbus some years ago. Agreement is within $\pm 5\%$.

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Electrical Resistivity of UO_2 . The apparatus that had been used to obtain information concerning the effect of atmosphere on the electrical resistivity of UO_2 at temperatures to 1600 C was disassembled, and a portion of one sample (MR-5) is being analyzed. The remainder of the specimen is being prepared for measurements at higher temperatures.

The electrical resistivity of UO_2 in the higher temperature range (to ~ 2500 C) was measured in a mixture of argon + 8% hydrogen, as in previous experiments. Single crystal UO_2 (MR-1) was successfully heated by self-resistance using 60 cps alternating current. Radiation and conduction losses caused in a temperature gradient across the specimen, thus optical pyrometer surface temperature measurements were low. An attempt was made to compensate the surface cooling effect by controlling heat input from an auxiliary strip heater. Voltage probes were inserted between the specimen and tungsten electrodes.

Results from the two methods of measurements agree very well. Reproducible data are obtained in an A - 8% H_2 atmosphere for temperatures between 800 C and 1500 C. In the temperature range above ~ 1500 C, the $\ln \rho$ vs $1/T$ relationship becomes nearly linear, with an activation energy ~ 1.9 eV. An extrapolation of the curve to 2800 C provides a resistivity value of $\sim 3.5 \times 10^{-3}$ ohm-cm, slightly higher than a previously reported value of 1×10^{-3} ohm-cm obtained with near molten UO_2 .

Thermal Expansion of Single Crystal UO_2 . A technique was developed for thermal expansion measurements to cryogenic temperatures, using a dilatometric device made of fused silica insulated with fiberglass cloth. A high conductivity layer of heavy aluminum foil is imposed between the fiber glass wrapping and the fused silica sample holder tube to distribute the heat evenly along the sample holder. The assembly is cooled in a liquid nitrogen bath and allowed to warm slowly to room temperature in a pre-chilled Dewar flask. Slow heating rates can be obtained in this manner, resulting in highly accurate data.

The coefficient of thermal expansion of a UO_2 single crystal between -190 C and +25 C was observed to be 7.242×10^{-6} C $^{-1}$. The averaged coefficient of thermal expansion of single crystal UO_2 between -190 C and +1550 C is 11.28×10^{-6} C $^{-1}$.

Electrodifusion of Oxygen in UO_2 . Preliminary experiments demonstrated the diffusion of oxygen ions in UO_2 by application of a small electric potential. A direct current electric potential (approximately 2 V/cm) across a UO_2 single crystal at 1600 C for 11 hours resulted in mass transport of oxygen to the positive end and precipitation

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uranium throughout the UO_2 near the negative end. The redistribution of oxygen in the electric potential gradient is evidence of ionic diffusion in UO_2 .

Fractography of Irradiated UO_2 Single Crystals. Continuing the evaluation of the effects of low level irradiation on UO_2 , the fractography of several arc-fused single crystals is being studied by electron microscopy replica methods. Two wafer specimens cut previously from one crystal were irradiated in a Hanford reactor experimental facility to 2×10^{15} f/cc and 8×10^{15} f/cc. These, and two nonirradiated control specimens from adjacent positions in the same crystal, were fractured and immediately replicated for microscopy.

Initial examination shows:

- (1) smooth pore-free surfaces in the nonirradiated specimens, a variety of fracture modes in each surface probably related to stresses produced in the crystal during solidification from the melt, and
- (2) evidence of incipient internal porosity after irradiation, particularly in the higher exposure specimens.

The mode of fracture appears to be related to mechanisms suggested earlier: point defects apparently accumulate during irradiation until agglomeration occurs to form visible pores.

Cermet Fuel Pin Fabrication. Four stainless steel-30 vol% UO_2 cermet extrusions were made to establish the processing requirements for fabricating four stainless steel- PuO_2 and one stainless steel-enriched UO_2 cermet fuel pins for EBR-II irradiation testing. These pins were fabricated by pneumatic impaction of cermet billets followed by hot extrusion to final size at a reduction ratio of 7.5/1. Dimensional variations of as-extruded pins are ± 0.007 -inch on the pin OD and ± 0.003 -inch on clad thickness.

Based on the above variations, the test fuel pins for EBR-II will be assembled for charging in the as-extruded condition. As-extruded pins with associated dimensional variation was judged superior to cold swaged pins with much lower dimensional variation for purposes of this test since minimum particle damage (no stringering and at most 5% fracturing) exists in the as-extruded pins.

Preparation of Rounded Fuel Particles. The feasibility of using a "jet grinder" to produce high density, rounded particles from angular UO_2 (or PuO_2) particles was demonstrated. The jet grinder consists basically of an alundum tube into which streams of air are introduced

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circumferentially through nozzles located in the bottom plate of the housing. The material to be ground is picked up in the circumferential air flow and "swirled" around the inside of the alundum tube. After only 10 minutes of grinding, the corners and edges of the angular particles are rounded; however, flat faces remain. As grinding time is increased, the particles become more rounded. After 30 minutes of grinding, the particles appear to be sufficiently round to provide the desired features for cermet use that are obtained with spherical particles.

The roundness of particles as a function of grinding time for both pneumatically impacted and arc-fused UO_2 was determined. It appears that rounder particles are obtained with arc-fused UO_2 than with impacted UO_2 ; however, this difference may be due to an operating variable rather than a difference in grinding characteristics of the two materials.

Air flow (grinding rate) was increased for the arc-fused UO_2 ground 40 minutes and 53 minutes. The high degree of spheroidicity obtained in these runs was at the expense of yield--only about 10% yield of the -60 +100 mesh size fraction was obtained.

Studies are under way to determine optimum feed material particle size, air flow, and grinding time to obtain maximum yield of particles of desired roundness.

Pneumatic Impaction of BeO and BeO-Containing Fuels. Further metallographic examination of a 99% TD pneumatically impacted BeO pellet (impacted at 1700 C, 274,000 psi) revealed an average grain size of ~10 microns. Previously observed larger "particles" (~500 microns), exhibiting microstructural characteristics similar to lower density BeO pellets (~85% TD), were found to consist of many small (<2 microns) BeO particles. This effect is tentatively attributed to agglomeration of the submicron BeO particles prior to impaction.

BeO-25 vol% UO_2 pellets were pneumatically impacted using the graphite can/MgO matrix technique. Preliminary examination of these pellets indicated that (1) the pellets were intact, with the exception of crazing of the surface; (2) impacted densities may be at least 95% TD; and (3) some UO_2 -graphite interaction may have occurred, even though contact time at temperature was minimized (<15 minutes).

Sodium Bonding Facility. Major design work on the sodium bonding facility (Phase 1) is complete. Some purchased parts and fabricated parts have been received. The few remaining components to be sent off-site for fabrication will be in process next month.

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Materials and Information Exchange. Six single crystal specimens of UO_2 and one specimen of pneumatically impacted UO_2 were prepared, characterized, and delivered during a visit to Argonne National Laboratory. Specimens are to be used in basic studies of thermal diffusivity and the chemistry of the U-O-W system. In addition, UO_2 crystals were prepared for shipment to Chalk River, AECL, and to Watson Research Center, IBM Corporation.

To date, more than 380 single crystal UO_2 specimens of various geometries have been prepared and exchanged with domestic and foreign research investigators for basic studies and information exchanges.

4. Basic Swelling Studies

Irradiation Program. The third controlled temperature-pressure capsule, containing six high purity uranium, two U + Fe-Al and two U + Fe-Si specimens, operated successfully in a reactor at 625 C and 1000 psi to goal exposure and was discharged. It was shipped to the Radiometallurgy Laboratory where it is being disassembled to explore the effect of pressure at specified temperature on the behavior of fissionable metals. A fourth controlled temperature-pressure capsule was charged in a reactor and is operating successfully at 450 C and 500 psi.

The fifth and sixth controlled temperature-pressure capsules have been assembled in a tandem array to run concurrently in a reactor at 500 psi and 550 C and 625 C, respectively. Each capsule contains six high purity uranium specimens, two U + Fe-Al specimens and two U + Fe-Si specimens having several heat treatments.

Assembly of two general swelling capsules was initiated to investigate the effect of burnup (0.1 and 0.2 at.%) at 700 C (beta phase) on the behavior of fissionable metals. Each capsule will contain high purity uranium specimens and uranium alloy specimens with various metallurgical histories.

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. Unirradiated control specimens of these alloys and Hastelloy C, Hastelloy R-235, and TD Nickel were tested at

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room temperature, 300 C, and 650 C. Data from these two series of tests are being compared to determine the effects of irradiation temperature and exposure on mechanical properties.

A temperature-controlled, liquid-metal, convection capsule has been designed for insertion into the ETR to determine the effects of irradiation in a liquid metal environment. A bench model of this capsule was fabricated and was successfully operated at a maximum temperature of 500 C. Loss of convection in the capsule was observed after about 80 hours of operation due to the influx of liquid metal into the vacuum annulus surrounding the capsule heater. Examination of the capsule showed that a defective weld was the cause of failure.

In-Reactor Measurements of Mechanical Properties

The status of three active in-reactor creep experiments is reported below with reference to test materials:

Annealed 304 SS. The 525 C, 30,000 psi creep test exhibits a rate of 1.1×10^{-6} in/in/hr after 1000 hours of testing. This rate is lower than expected and considerably lower than the ex-reactor creep rate of 5×10^{-6} in/in/hr.

A 600 C, 10,000 psi creep test was terminated this month after 1700 hours of testing at which time the creep rate was 4×10^{-6} in/in/hr. Two ex-reactor control tests exhibited comparable rates of 2.1 and 3.6×10^{-6} in/in/hr. Test termination was preparatory to conducting creep activation energy studies. These measurements will be made at the same conditions of temperature and stress just prior to and just after reactor shutdown.

Annealed Zr-2. The PRTR in-reactor creep test, containing an annealed, longitudinal, Zr-2 specimen taken from plate material, was started this month. Test conditions are 315 C and 15,000 psi. Due to an extended, unscheduled reactor outage, the test was stopped shortly after specimen loading while creep was still in the primary stage.

Irradiation Effects in Structural Materials

The purpose of this program is to investigate the combined effects of irradiation and reactor environment on the mechanical properties of structural materials. Special attention will be given to the determination of mechanical property changes produced in metals by irradiation at elevated temperatures.

Tensile tests were conducted at both room temperature and 550 F (290 C) on BONUS transition-joint specimens irradiated in the G-7 loop to

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exposures between 4.0 and 6.0×10^{20} nvt (>1 Mev). These were the last in a series of tests designed to determine the strength and integrity of transition joints after exposure in a nuclear environment similar to that of the BONUS Reactor. An additional reduction in ductility beyond that observed in earlier tests at lower exposures was noted at both test temperatures. This loss of ductility, which results in the failure of the Zircaloy-2 portion of the specimen, is due to defects introduced by the neutron flux and by the formation of hydrides. Preliminary results indicate that the rate of hydride formation in these specimens is far greater than normal. However, since the specimens exhibited very low ductility in the as-received condition, it is difficult to assign the cause for additional embrittlement to either hydride formation or neutron irradiation.

The specimens investigated were taken from two types of transition joints supplied by different vendors. These two types varied considerably in the amount of hydrogen accumulated in the Zircaloy-2 during exposure, and these differences were reflected in the strength and ductility of the specimens. A topical report summarizing the results of this cooperative study in support of the Advanced BONUS Core Development Program is being prepared.

A study on the effects of geometric variables on the calibration and fracture behavior of DCB, fracture-toughness specimens has been completed. The variables included in this study were thickness, beam depth, side groove depth, and side groove root radius. In addition to determining the compliance calibrations for various combinations of these variables, the fracture toughness was also measured. These data have proved invaluable for specimen design. A paper concerning this investigation was presented to the ASTM E-24, fracture subcommittee.

Irradiation Effects on Nickel-Base Alloys

The purpose of this program is to determine the effects of modified microstructures on the irradiation stability of nickel-base alloys. Structural modifications are made by pre-irradiation thermal and/or mechanical treatments and are screened by tensile tests, stress-rupture tests, and metallographic examination.

Analyses of the results of tensile tests at room temperature and 1350 F (730 C) on Hastelloy X-280 specimens irradiated at 540 F (285 C) to 1×10^{20} nvt and representing eight thermal/mechanical pre-irradiation treatments have continued. When compared with a standard treatment, some treatments showed improved high temperature ductility while maintaining strength after irradiation; and, in general, susceptibility to radiation damage was lessened. Another treatment results in

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an 84% increase in yield strength with little reduction in ductility at high temperature and a 63% increase in yield strength and retention of 18% ductility at room temperature.

Metallographic studies of the tensile specimens, buttonheads and fracture surfaces are in progress. Stress-rupture tests on irradiated specimens are to be initiated next month at Battelle-Columbus.

Eighteen specimens of Hastelloy X-280 and Inconel X-750 with experimental heat treatments are being irradiated through Cycles 72 and 73 at 540 F (285 C) in the ETR. Additional specimens are being prepared for 1300 F (705 C) irradiations.

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.

Additional analysis of the data from double strain rate change tests showed that failure of the assumption that

$$\left(\frac{\Delta \ln \dot{\gamma}}{\Delta \tau}\right)_T = \left(\frac{d \ln \dot{\gamma}}{d \tau}\right)_T = \frac{V^*}{KT}$$

In regions where these quantities are changing rapidly leads to erroneous values of V^* . The most serious consequence of this error is the failure of the activation energy to follow a linear temperature dependence in the low stress regions (200-300 K in iron), when H is evaluated from experimental data by the relationship

$$H = -TV^* \left(\frac{\Delta \tau}{\Delta T}\right)_Y$$

This error has been universally overlooked by workers in the field of thermally activated deformation and has led many to conclude that another mechanism becomes controlling in the region where H ceases to be linear with T . By using the double strain rate change data, a back extrapolation could be made to find an approximate value of the tangent to the $\ln \dot{\gamma} \tau^*$ curve at the initial value of τ^* rather than the secant to the curve obtained when one has only a single value of $\Delta \tau$ to use. Re-evaluation of the H - T curve using these refined values of activation energy showed the relation was linear to the highest temperatures at which thermal activation occurred.

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This result has further implications in discussing the mechanism of deformation in Ferrovac E and BMI iron which are very similar but have slightly different values of H_0 , the activation energy at zero stress. It appears that the mechanism in this range may be the nucleation of double kinks to surmount the Peierls stress hill, which controls at lower temperatures. The mechanism is modified in some way to make nucleation easier when impurities and/or defect clusters are present.

Combining the observations from these studies with the calculations of Makin on long range forces from dislocation loops, the following tentative model of radiation damage is proposed. Additional experiments at higher exposures will be required to confirm it.

Defect clusters or loops affect dislocation motion by both long range and short range forces. The short range forces influence the effective stress, τ^* and the long range forces influence the internal stress, τ_i . The proportion of these two effects changes with the size of the loops with the short range effects decreasing and the long range increasing as the loops grow. The loss of defects to other sinks makes it difficult to confirm this prediction. A simple comparison of yield stress before and after irradiation is not an adequate test of this effect either, since a modification of mechanism may, for example, cause an increase in τ^* and a decrease in τ_i which would have the same yield stress as an unirradiated sample.

The significance of these results lies in considering ways to improve the radiation resistance of a material. The deterioration of ductility is due to localized deformation. The larger loops have the longest range interaction with dislocations. Therefore, a dislocation moving through such material will sweep up loops over a wide zone, weakening the material in this zone and making it subject to localized deformation. To reduce this tendency, which is greater for the larger loops, one must inhibit the growth of large loops either by nucleating more small clusters or by additives which would relieve the strain energy of individual defects and reduce their tendency to cluster.

ATR Gas Loop Studies

Model Loop Operation. The model gas loop was operated to determine the impurity level that could be maintained during a high temperature run. The loop eventually was shut down for repair of leaks in nonwelded connections and seals.

Total operating time on the gas bearing compressor is 481 hours, with 115 starts.

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Columbium Sleeve Leak Test. The installation of the test fixture has been completed and room temperature runs have been started on the prototypical ATR gas seal for the in-reactor tube. The nominal diametral clearance between the columbium sleeve and the Invar seal is 0.010 inch, which is the minimum allowed under the design specifications. When this test run is completed, the test fixture will be modified to give the maximum allowable clearance, and a second set of data will be gathered.

Model Gas Loop Heater Design and Piping Modifications. Foil insulation sections for the model loop concentric heater are 60% complete. The first attempts at making sections of foil insulation indicated that original designs of 30 layers per inch provided no strength; the foil would not support even an inner liner. Increasing the density of foil to 60 layers per inch helped support the inner liner but was not desirable from a conductivity standpoint. A satisfactory design was made with 30 layers per inch of foil by inserting pins, made from Haynes-25 welding rod, through the outer liner and foil and either butted against or welded to the inner liner. The pins were then welded to the outer liner.

No additional work has been done this month on the detail drawings of the pipe modifications for the model loop.

Four-Way Valves. Leakage was detected in the Barksdale four-way valves used on the room temperature molecular sieve beds and on the low temperature adsorption beds. Helium was leaking into the valve casing and between the high pressure side and the side being regenerated. The "shear" disc and sealing rings were scratched. These were hand lapped and given a final polish on the Syntron Polisher in the Metallography Laboratory. The Viton-A O-rings and Teflon backup rings were repacked with Dow Corning silicone grease and the sliding surfaces given a coating of molycote before reassembly. The valves have operated satisfactorily for 24 hours following this renovation.

Temperature Controllers. The temperature controllers for regeneration of the dual charcoal and molecular sieve low temperature adsorption beds were found to be faulty. The #1 Bed controller was about 200 F (110 C) low in its calibration, resulting in too low a temperature for proper regeneration. The #2 Bed was stuck in the "on" position, resulting in a very high temperature of regeneration. No damage was done to the charcoal or molecular sieve material in the bed, but the foamed polyurethane insulation surrounding the dewar flask was charred.

The #1 Bed controller has been recalibrated and the #2 Bed controller has been replaced with a calibrated controller. Test thermocouples for checking bed temperature have been installed on both beds.

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Helium Relief Valve Seat Leakage Evaluation. The Consolidated Relief Valve #1975T, equipped with a Vitan-A O-ring seal has been evaluated. The valve was set to relieve at 500 psi. No leakage was detected before exceeding the set pressure. Following operation of the valve, either manually or by over-pressure, the valve reseated at 480 psi and no leakage was detected when the pressure returned to 500 psi.

The test was conducted at both 75 F (25 C) and 150 F (65 C). The leak detector used was a Consolidated Electrodynamics Corporation Helium Leak Detector #24-120A, operating on the most sensitive scale. This valve is considered acceptable for use in the ATR Gas Loop.

Moisture Analysis. A temperature dependent change in the moisture analysis of a tank of helium, with accurately known impurities, was noted from cool morning to warm afternoon conditions. The moisture analysis stabilized at 6 ppm at 60 F (15 C) and rose to 8 ppm at 80 F (30 C).

It was further noted that the sintered stainless steel filter used to prevent small particles from entering the moisture detection cells holds large quantities of water and causes a long delay to stabilize the moisture analysis indication. The filter will be heated to about 350 F (175 C) to determine if this increases the reliability of the analysis and speeds up the response time.

Corrosion Studies. A single oxidation test on Hastelloy X-280 and Haynes Alloy 25 has been performed in a double-contained system previously described. This test was primarily conducted to establish superalloy oxidation - evaporation behavior in low oxidant partial pressures. Two coupon samples (0.75 inch x 0.50 inch) of each alloy were placed in the reaction zone. A replenished dry air atmosphere was established and maintained at 3×10^{-4} torr. The temperature was then raised to 1120 C (605 C). The weight change due to both evaporation and oxidation for these coupons at the end of a 45-hour exposure is listed below.

	Weight Change (mg/cm ²)	Retained Oxide (mg/cm ²)	Oxide Thickness (mils)	Total Metal Penetration (mils)
Hastelloy X-280	-4.8	Not Determined	--	>0.23
Haynes Alloy 25	-2.8	Not Determined	--	>0.12
Hastelloy X-280	-5.1	0.18	0.02	0.26
Haynes Alloy	-2.5	0.36	0.05	0.16

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Retained oxide was determined by descaling the samples. The sample weight loss was assumed to be due primarily to evaporation because (1) the sample holder was coated almost uniformly with a green oxide, indicating either gas phase oxidation of the evaporating metal or deposition of metal on the sample holder and subsequent oxidation, and (2) no visual evidence of spalling of the oxide film formed on each of the four coupons.

The results indicate that evaporation in near vacuum conditions will proceed until sufficient oxidant can diffuse through the evaporating medium and form solid oxide on the metal surfaces. One contributing factor which could have enhanced evaporation in this case was the build-up of nitrogen near the sample surface. Future tests will consider pure oxygen as the contaminant with the intent of reconciling evaporation-oxidation phenomena at low reactive gas pressures.

Boiling Burnout Studies for the ATR. Heat transfer experiments were performed on an electrically heated test section which was a full-length simulation of a typical rectangular coolant channel of the Advanced Test Reactor (ATR). The test section coolant channel was 2.028 inches wide, 0.070 inch thick, and 49.5 inches long. Both 2-inch wide sides of the test section were heated along a 48-inch length by passing direct current through 1.90-inch wide copper-nickel bars which were brazed to the 0.010-inch thick stainless steel forming the flow channel. These Cu-Ni bars were shaped longitudinally to produce a chopped cosine axial heat flux distribution with a 1.41 peak-to-average ratio. Also, these bars were tapered laterally to produce a nonuniform lateral flux distribution with a value of 1.25 peak-to-average. These conditions were selected to simulate a pair of fuel plates in the ATR in a region of skewed nuclear flux.

The hydraulic performance curves for this model were determined at five different power input rates which produced local heat fluxes up to 4 million Btu/hr-ft². In addition, three runs were conducted in which the power input rate was increased until flow instability occurred and film boiling conditions were initiated within the test section.

Hydraulic demand characteristics of the channel were determined by holding power input and inlet temperature constant while the flow rate was reduced in small steps. Test section pressure drop was recorded for each step. Flow reductions were continued until the channel pressure drop began to increase due to boiling. These hydraulic demand curves were obtained with an inlet temperature of 130 F, mass velocities of about 9 million lb/hr-ft², and peak local heat fluxes of about 2.1, 3.3, and 4.0 million Btu/hr-ft². In addition, curves were obtained for two very low heat fluxes which simulate reactor shutdown conditions.

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The flow instability experiments were performed in a piping system which simulated the parallel operation of 10 coolant channels. With this system the test section pressure drop remained nearly constant even when the test section flow rate varied because of boiling and two-phase flow.

Three different instability tests were made during which the power input was increased until flow instability and film boiling were initiated in the test section. Peak local heat fluxes for these runs were about 3.5 million Btu/hr-ft².

The data from these experiments show that coolant mixing across the 2-inch width of the channel was far from complete. Three thermocouples positioned in the coolant channel near the outlet end indicated cross-channel temperature differences of 70 to 100 F. In addition, the inflection points in the hydraulic demand curves were found to occur before the outlet coolant reached saturation temperature.

These data obtained with the nonuniform heat flux test section will be compared with those obtained from a previous test section of uniform lateral heat flux. This comparison should aid in determining appropriate hot spot factors for Advanced Test Reactor.

6. Nuclear Graphite Studies

EGCR Graphite Irradiation. The program for the long term irradiation of EGCR graphite is progressing satisfactorily. The H-3-10 capsule has successfully completed three cycles of irradiation and the maximum neutron exposure is estimated to be 1.75×10^{22} nvt, $E > 0.18$ MeV.

High Temperature Graphite Irradiations. The second capsule, GEH-13-11, in the series of high temperature irradiations of graphite is operating in its fourth cycle. Detailed checks during the past shutdown period showed that in most cases the electrical resistance across the thermocouples had increased. Upon reactor startup, three thermocouples continue to function properly while the remainder have all failed. From the information presently available it appears that one leg of the thermocouples may have carburized with a resultant change in calibration. To date, none of the thermocouples have broken or otherwise opened up.

In-Reactor Strain to Fracture. The initial results of the in-reactor strain-to-fracture test on 14 transverse tensile specimens of EGCR-type AGOT graphite show a maximum strain of $\sim 0.25\%$ at an exposure of 1×10^{20} n/cm² ($E > 0.18$ MeV) and a temperature of 625 ± 25 C. This compares with unirradiated strains to fracture of $\sim 0.15\%$. Further tests

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to determine the amount of irradiation-induced plasticity at different strain rates are in progress.

Effects of Additives on Graphite Contraction. Speer Contract DDR-118 samples have been discharged at a total exposure of 6400 Mwd/At_K. The samples were irradiated in KW Reactor at a temperature of 600 ± 100 C. The sample set utilized TSX, as the base graphite, with the following chemical additives:

0.5, 1.0, 2.0, and 4.0%, Fe₂O₃; 1.0% Fe₂O₃ (purified),
 1.0% FeO, 1.0% Fe₃O₄, 1.0%, FeS, 1.0% FeSiO₃,
 1.0% Fe, 1.0% Cr₂O₃, 1.0% CrCl₃·6H₂O,
 1.0% CuO; 1.0% V₂O₃, 1.0% ZnO,
 1.0% Ni₂O₃, 1.0% Al₄C₃.

All additives were technical grade with the exception of the 1.0% purified Fe₂O₃. During the processing to graphitization temperature the additives are volatilized leaving a graphite of nuclear purity. Based on dimensional stability of both orientations the most promising additives, to this exposure, are itemized below:

Sample	Length Change % (6403 Mwd/At _K)		Rate of length change, % ΔL/L per 1000 Mwd/At _K (4203 to 6403 Mwd/At _K)	
	⊥		⊥	
Base graphite (no additive)	-0.034	-0.082	-0.0118	-0.0218
1.0% Cr ₂ O ₃	-0.019	-0.068	-0.0073	-0.0182
1.0% Cr ₂ Cl ₃ ·6H ₂ O	-0.018	-0.074	-0.0077	-0.0186
1.0% Al ₄ C ₃	-0.014	-0.074	-0.0068	-0.0177
1.0% V ₂ O ₃	-0.001	-0.068	-0.0059	-0.0191

Since the base material is one of the more dimensionally stable production-grade graphites, the lower contraction rates represent an appreciable gain. At this exposure no definite trends are evident on the coefficients of thermal expansion.

Replica Electron Microscopy of CSF and KCF Graphites. Length measurements were made in identical areas before and after irradiation on electron micrographs of CSF and KCF replicas with an optical comparator. The CSF sample was irradiated at 700 C to 1.1×10^{21} nvt, E > 0.18 MeV,

and the KCF sample was irradiated at 30 C to 1×10^{19} nvt, $E > 0.18$ MeV. The measurements were made over a distance of about 5 microns on the specimen (1/2 inch on 5000X photographs).

The CSF replicas were taken from a portion of a coke particle. Comparison of the CSF replicas showed that irradiation caused the direction normal to the Mrozowski cracks (i.e., the c-axis direction) to expand from 2 to 8%. Parallel to the cracks (a-direction) the coke particle contracted from zero to 5% depending on the location of the examined areas. In contrast, the KCF graphite irradiated at 30 C appeared to have an irradiation-induced length change independent of direction from -2% up to as much as +11% with no apparent correlation with the texture. The measurement error is about 2%.

Examination of photographs of two different CSF replicas of the same area after irradiation gave the same result. The variations observed in the c-axis direction could be due to differences in the angle at which the particles intersect the surface.

A pyrolytic graphite irradiated at 700 C to 8×10^{20} nvt (7000 Mwd/At), indicated 1.5% c-axis expansion and 0.6% a-axis contraction. The crystallite changes calculated by substituting thermal expansion and bulk length change data into the Simmons model indicate 0.5% c-axis expansion and 0.14% a-axis contraction for the CSF material. From the preceding numbers, it appears that the calculated crystallite changes underestimate the actual changes in the CSF coke particle. This difference might be explained by a binder effect, internal stresses, or may indicate that the CTE data cannot be used in calculating accommodation coefficients for irradiation-induced length changes. Another possible explanation is that more deformation occurs at the surface than in the bulk of the body.

Visual examination of the photomicrographs shows many cracks parallel with the layer planes in the CSF material, but cracks are not evident in the KCF graphite. Measurements with a protractor gave sheer displacements up to 6° between the before and after irradiation replicas of the CSF material, but significant sheer distortions were not evident between the before and after irradiation replicas of the KCF material. The cracks in the CSF material may have resulted in part from sheer displacements adjacent to the layer planes during irradiation, but definite sheer displacements immediately between adjacent cracked layer planes were not observed. These cracks could also be a surface effect, or they might have been due to tensile stresses developed from a-direction shrinkage.

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This latter possibility ought to be considered because the bulk material shows about a 0.2% contraction at this irradiation level. If the a-direction shrinkage is this dominant in causing property changes, then the procedure of using thermal expansion data for predicting irradiation-induced bulk length changes may not be accurate because a-direction thermal expansion changes are very small.

Inert-Gas-Sensitized Radiolysis of Carbon Monoxide-Water Vapor Mixtures. Previously the helium and argon sensitization of the radiolytic reaction of carbon monoxide with water vapor had been reported. Both inert gases sensitize the radiolysis very effectively and one of two possible mechanisms appear to be responsible for the sensitization:

- (1) Direct charge exchange

where X represents an inert gas atom.

- (2) Neutralization of inert-gas ions followed by energy transfer

Both He^+ and A^+ are sufficiently energetic to charge exchange with both CO and H_2O by an exothermic process. Likewise the energy-transfer process is also energetically feasible.

The possible sensitization of the radiolysis reaction by krypton and xenon has been studied. The ionization potential of krypton is slightly less than that of CO but greater than that of H_2O . Xenon, however, has an ionization potential smaller than both CO and H_2O . Krypton could, therefore, undergo charge exchange with H_2O but not with CO, and Xe would not be expected to undergo charge exchange with either CO or H_2O .

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We have studied the radiolysis of CO and H₂O in the presence of 75 mole % of both Kr and Xe. Both of these inert gases sensitize the radiolysis reaction very efficiently. The 100 ev yields (based on total absorbed energy) of H₂, CO₂, and CH₄ are the same as observed when no inert gas is added. Since direct charge exchange is unlikely with Xe⁺ for the reason given above, the results suggest that the energy exchange after neutralization is responsible for the sensitization at least in the case of xenon and perhaps also for the other inert gases.

Oxidation of Boronated Graphite. Some preliminary measurements of the rate of reaction of boronated graphite with water vapor have been performed. As anticipated, the weight changes observed are highly dependent on the previous oxidation history of the sample. The first sample initially contained a nominal 5 wt% boron (grey) but had received a previous apparent oxidation of about 5.7% in oxygen and hence contained appreciable B₂O₃. The following rates were found upon exposing this sample to helium containing approximately 6000 ppm H₂O and flowing at 150 cm³/min.

Run No.	Temp., °C	Apparent Oxidation Rate, hr ⁻¹
1	532	-2.99 x 10 ⁻⁴
2	528	-2.90 x 10 ⁻⁴
3	634	-2.35 x 10 ⁻⁴
4	730	-3.15 x 10 ⁻⁵

It is seen from the above table that the rate of weight loss decreased with time even though the temperature was increased. No H₂ was found at any temperature by a chromatographic analyses of the effluent gas. It is thus concluded that the observed weight losses were due entirely to the removal of B₂O₃ from the sample by H₂O. The rate of weight loss decreased with time as a consequence of decreasing B₂O₃ concentration. The absence of H₂ indicates that neither the graphite nor the contained boron carbide reacted with water vapor to a measurable extent under the experimental conditions employed.

The second sample of boronated graphite was identical with the first but had experienced no previous oxidation. The experimental conditions were the same as before. At 730 C, a very slow weight increase was observed and about 20 ppm H₂ appeared in the effluent gas. At 826 C, the rate of weight increase was appreciably faster and, initially, about 90 ppm H₂ were found in the effluent gas. After passing through a maximum, the sample weight eventually decreased at a constant rate and about 40 ppm H₂ were noted. Under none of these experimental

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conditions were any oxides of carbon observed. It thus appears that the water vapor did not react with the graphite but rather with the boron carbide as follows:

This reaction then accounts for the initial weight increase. The eventual weight loss occurred after the rate of B_2O_3 removal by H_2O exceeded the rate of B_2O_3 formation.

Because the reaction of boronated graphite with oxygen occurs at lower temperatures than the water-vapor reaction, future work will be concerned with the former reaction. Subsequently, the reaction of boronated graphite, both unoxidized and pre-oxidized, with water vapor will be investigated in greater detail.

7. Metallic Fuel Development

Irradiation of Thorium-Uranium Fuel Elements. The irradiation of three tubular Zircaloy-2 clad thorium - 2.5 wt% uranium - 1.0 wt% zirconium fuel elements continued successfully in the M-3 loop. Three fuel elements were not irradiated because the P-7 loop was used for another experiment. Current irradiation conditions are:

Loop	Core Temp., °C	Surface Heat Flux Btu/hr-ft ²	Int. Exp. Fissions/cm ³ (Mwd/ton)	Fuel Vol. Increase %Δ/V
M-3	500	6.5×10^5	4.0×10^{20} (11,750)	1.3

One fuel element from the P-7 loop which has attained an exposure of 1 at.% burnup will be intentionally defected and tested in the IRP loop at Hanford and then metallographically examined.

Platinum Coatings by Evaporation. Six copper-nickel alloy rods, one nickel rod, and one iron rod have been coated with platinum to a uniform thickness of 0.0012 inch using an electron beam evaporator. The rods were 3 inches long by 0.340 inch diameter. Surface preparation and substrate temperatures were critical in forming the thick, strongly-adhering deposits. Vacuums as low as 5×10^{-8} torr contributed greatly toward keeping the substrate clean during the course of the evaporations. Evaporation conditions were as follows: substrates were polished with dry 4/0 metallography paper immediately before putting them in vacuum. Rods were positioned 9 inches from the platinum source and were heated to 200 C for a minimum of four hours before each plating. The rods

were rotated 45° after each one hour of evaporation to insure a uniform coating and required a total of 8 hours of evaporation to coat all surfaces to the required thickness. The platinum was positioned in a water-cooled copper crucible and was heated with 4.2 KW of electron beam power. The coated rods will be used to make thin platinum honeycombs by extrusion.

8. Advanced Reactor Concept Studies

Nitride Fuel Cycle Studies. Since the previous reporting period, work has centered around generating a suitable cross section library to carry on analysis of the nitride fast reactor core. Considerable core physics information was obtained with an HFN data tape generated with HRG; criticality vs. enrichment, sodium void coefficient (partial and total), a carbide comparison, and an initiation of the Doppler temperature coefficient calculation. However, it has been observed that HRG data do not yield the neutron spectrum for the nitride fueled reactor one would expect. The HRG data calculated the peak flux to occur at 150 kev compared to previously reported peak fluxes of about 400 kev. Comparing HRG U-238 cross sections against GAM U-238 cross sections, it was found that significant differences occur in the energy range 300 kev to 1 Mev. A special HRG data tape has been generated containing GAM U-238 cross sections. With this special tape, new HFN data have been generated incorporating the GAM U-238. The new HRG calculated spectrum is harder and appears to be suitable for the physics calculations. A new HFN data tape is being generated and core physics calculations will be continued.

Programming of a heat-transfer parameter survey of nitride-core fuel pins was largely completed during the month. Variables included are pin size, heat generation rate, sodium and gas (nitrogen) bonding of fuel to clad, and the use of stainless steel (19-9 DL) and niobium clad. Results of the calculations are expected before the end of June and should aid in selection of optimum fuel design for the nitride fueled core.

9. Upstream Boiling Burnout

Recently obtained data were analyzed to study the relationship between the upstream boiling burnout phenomenon and the pressure drop behavior. These data were obtained on an electrically heated test section consisting of an Inconel tube 6-ft long and 0.435-inch ID. This test section was cooled by water flowing through its center with a mass velocity of 7×10^6 lb/hr-ft² and a pressure of 1500 psig. The following observations were made:

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- (a) When the outlet enthalpy was below saturation (614 Btu/lb), the observed pressure drop exhibited only a slight dependence on heat flux.
- (b) At higher outlet enthalpies up to 655 Btu/lb (550 Btu/lb inlet), the pressure drop increased steadily with heat flux up to the boiling burnout condition. In these cases the location of the burnout was observed to occur at the downstream end of the test section, and the burnout heat flux decreased as inlet enthalpy increased.
- (c) At outlet enthalpies of 675 to 725 Btu/lb (575 to 625 inlet), the pressure drop increased with heat flux until a maximum was reached. Then the pressure drop decreased with increasing heat flux until the boiling burnout condition was reached. Here, the burnout occurred at an upstream location on the test section, and the burnout heat flux was observed to be nearly independent of inlet enthalpy.
- (d) At outlet enthalpies of 770 to 810 Btu/lb (650 to 675 Btu/lb), the pressure drop was observed to go through an inflection as heat flux was increased. That is, it first increased, then decreased (or remained constant), and finally increased with increasing heat flux until boiling burnout conditions were reached. Here, the burnout was observed to occur near the upstream end of the test section, and the burnout heat flux increased with increasing inlet enthalpy.

In runs with no heat flux, the pressure drop increased linearly with enthalpy up to at least 700 Btu/lb. Since the maxima in the pressure drop-heat flux data were observed to occur at outlet enthalpies below 700 Btu/lb, the reduction in pressure drop must not be a function of enthalpy alone but must be also dependent on heat flux.

These data suggest that a transition in flow pattern occurs at some enthalpy and heat flux to a new flow pattern with a lower pressure drop. This flow pattern change could possibly explain the minimum in the graph of burnout heat flux vs. enthalpy which appears to be responsible for the upstream burnout phenomenon. Additional experiments are being planned to study these effects further.

10. Fast Flux Test Facility Studies

A review in depth of some of the more basic design concepts of the FFTF was made by AEC Headquarters and RLOO-AEC June 29 and 30. Results of this meeting are being transmitted via the meeting minutes.

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With concurrence of the AEC, issuance of the FFTF Conceptual Design Study report was deferred to August 25, 1965.

No additional consulting services were procured during the month. Procurement Directive RL00-65-5 in the amount of \$15,000 for Argonne National Laboratory Services was cancelled by mutual consent.

Preliminary PERT charts were prepared for the majority of the FFTF systems. From these charts take-offs were made on R&D requirements, costs, and schedules for the total FFTF program.

The conceptual design of the FFTF was continued vigorously during the month, and cost estimating is in progress. Certain items of particular interest relating to R&D are presented below:

Driver Fuel R&D. From the 6 kilograms of impacted UO_2 -20% PuO_2 fuel which were sent to ANL for EBR-II irradiation tests, some 36 pins will be fabricated. About 12 pins in each of three subassemblies will be irradiated. The subassemblies are tentatively scheduled to attain the following burnups: 5% (45,000 Mwd/ton), 10% (90,000 Mwd/ton), and 15% (130,000 Mwd/ton). If it is assumed that the EBR-II operates at 45 Mw and with a plant factor of 50%, then it takes about one year to reach 5% burnup.

The cermet pins for irradiation tests in one of the ANL subassemblies in EBR-II are scheduled for shipment to ANL in August 1965. The stainless steel clad, stainless steel matrix cermet pins are of three types: two pins contain 30 vol% PuO_2 , two pins 20 vol% PuO_2 , and one pin 20 vol% UO_2 . The pins are to be about 0.76 cm (0.3 inch) in diameter and 36 cm (14 inches) long and will each be irradiated in static sodium. All pins are to be placed in one subassembly which is scheduled to attain a burnup of 2% (takes about 130 calendar days). Desired centerline temperature in the 30 vol% PuO_2 pins is about 680 C (1250 F).

During the month the 8-pin cluster was discharged from the ETR. The burnup of the cermet pins is 5-6%. The pins are 0.64 cm (0.25 inch) in diameter and 30 cm (12 inches) long and have stainless steel (304L) cladding and matrices. The ceramic loadings vary from 20 to 30 vol% and consist of PuO_2 , $\beta\text{-Pu}_2\text{O}_3$, and UO_2 fully enriched). Estimated centerline temperatures were as high as 970 C (1780 F).

The last set of cermet capsules in the MTR are due out in July 1965. The capsules have stainless steel (304L) cladding and matrices. Both contain 20 vol% PuO_2 . The 82% dense and 97% dense cermets are operating with estimated centerline temperatures of 1400 C and 1200 C, respectively.

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The feasibility of making "rounded-off" fuel particles by a mechanical or jet grinding technique was demonstrated. The technique was applied to both high and low density UO_2 particles with no apparent changes in the fuel particle properties as a result of the processing. The equipment is being modified for glove box use and tests with PuO_2 particles.

Engineering Test Facility. Preliminary flow diagrams and equipment arrangement drawings for the Engineering Test Facility were completed. Two plans for the main system test loop are being studied. Both plans provide reactor core and environment simulation. One plan is a single loop system having flow and heating conditions of subassembly capabilities. The other plan provides flow rate and partial heat capacity of one reactor heat transfer loop including the secondary and tertiary systems.

Cursory calculations of piping thermal stresses were made. Pipe wall thickness in sizes of interest calculated to meet code allowable stresses appear to become highly thermally stressed upon ramp temperature as might result from a scram simulation. More detailed examination of this problem is expected.

Metallurgy. Items of metallurgical research and development in support of the design, construction, and operation of the FFTF have been formalized and written in preparation for inclusion into Volume III of the report. These items include research and development required in the areas of irradiation effects on mechanical properties of structural materials, corrosion, materials compatibility, reactor component testing and evaluation, and materials properties. Major items of capital equipment, manpower costs, and associated schedules have also been determined.

A report has been completed which will form the basis for neutron exposure criteria as well as outline the radial flux distribution in Core Concept III. Use of these data will facilitate the selection of proper dimensions of the core and vessel components of the reactor.

Chemical Metallurgy. A number of specific programs of chemical metallurgy research related to the FFTF were proposed. These are summarized below by title and proposed site:

- (1) Coolant Impurity Study - Cooperative program with all participating FFTF sites, particularly BNW, GE-APED, AI, and ORNL.
- (2) Hot Cell Analytical Methods - BNW Chemistry Department & Oak Ridge National Laboratory should cooperate.

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- (3) Sodium Sampler - AI, hardware development. AI has existing technology in sampling.
- (4) Self-Welding of Stainless Steel Process Tubes - Battelle-Columbus-
a) best sodium friction studies, b) B-C has existing equipment.
- (5) Fuel Failure Mechanisms - BNW.
- (6) Fission Product Handling - BNW.
- (7) Corrosion Surveillance Tab Holders - AI - experience in Hallam and SRE tab holder development.
- (8) Ferritic Steel for Reactor Secondary - possibly AI - much past work in this area.
- (9) In-Reactor Corrosion Effects - BNW-ORNL-ANL - cooperative program with irradiation effects on materials.
- (10) Sodium Vapor Trap - ANL - good prospective design; AI - past work for Hallam and SRE.
- (11) Fretting of Fuel Elements in Process Tube - AI - primarily requires flow and vibration tests.
- (12) Cover-Gas-Sodium Interactions - BNW - broad background in gas chromatography.
- (13) Mass Transfer Studies - Loop Scale - GE-APED - redirect current loop construction to FFTF parameters.
- (14) Mass Transport of Radioactivity to Out-of-Reactor Piping - BNW.
- (15) On-Line Instrumentation - Quantitative Detection of Impurities - ceramics research at GERL needed. Development of O, H, and N analyzers at BNW. Consultation and R&D with ORNL analytical department. Testing of developed instruments by APDA.

Core Stress Analysis. A computer program has been written for a 2-region plate structure. The central region will be the grid type structure formed by the holes containing the reactor tubes. Partial uniform loads, concentrated ring loads, concentrated ring moments, mean temperature distributions, and temperature gradients are accounted for. The degree of edge restraint on the outer edge can be specified. This solution should handle cases for the top grid support, bottom grid support, and reactor cover. The particular loadings mentioned above should be

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sufficient to cover all those encountered by these structures. The program is being debugged and check cases are being run.

Fuel Handling. The final report has been received from Atomics International for their conceptual design of a Fuel Handling Machine for FFTF. The report lists the following four areas of development-testing which covers a period of 30 months from receipt of funds to completion of the proof testing of the actual Fuel Handling Machine: (1) Sub-component testing, (2) General system testing in air, (3) General system testing in sodium, and (4) Proof test with actual FHM. Their estimate of total R&D effort is \$1,161,762 with an additional \$1,499,000 for design, fabrication, assembly, knockdown, crating, shipping to Hanford, and erection. The cost of the proof-test facility is estimated to be \$371,000.

Fuel Element Cleaning. Review of Fermi fuel cleaning procedure has indicated that the fuel element is cooled with argon with dry steam gradually introduced to remove the sodium. At Hallam the fuel element is cleaned using steam, water, nitrogen, vacuum, and helium in a sequence that yielded the most effective cleaning.

While it could be assumed that either of the above mentioned cleaning techniques would be satisfactory for FFTF, the higher decay heat output of FFTF elements indicates that other cleaning techniques should be investigated. One method that appears to satisfy the criteria for high cooling capability is a kerosene coolant loop with Cellosolve (ethylene glycol monoethyl ether) introduced to remove the sodium.

Leak-Tight Joint for Sodium Service. During refueling the fuel element must remain under sodium at all times. This can be accomplished in one of two ways. Either place the fuel element in a containment pot filled with sodium or flood the lower end of the FHM with sodium. The first method appears to be the most reliable and safe way to transfer fuel; however, it complicates the reactor tube configuration and lengthens the reactor tube approximately 10 feet. As there is considerable savings involved in reducing the length of the reactor tube (and reactor and building), it is believed that some R&D effort should be expended to investigate a leak-tight joint for sodium service.

Significant Letters and Reports. The listing below gives significant reports issued as part of the FFTF program activities:

Trip Report - Atomics International, Atomic Power Development Associates, EBR-II, Hallam Nuclear Power Facility, by R. V. Dulin, K. L. Young, K. D. Hayden, L. D. Perrigo, R. A. Thiede, June 9, 1965.

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Trip Report - NTH Products, Inc., El Cajon, California, by
G. S. Allison, June 14, 1965.

11. 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 and rhenium.

Analysis of molybdenum foils which were prestrained, irradiated, and deformed approximately 2% confirm the conclusion reached on previous prestrained samples. The pre-existing dislocations are strongly pinned during irradiation and do not act as dislocation sources during subsequent deformation. Some prestrained and irradiated molybdenum has been annealed at 600 C for two hours to establish the effectiveness of existing dislocations as sinks for point defects. At this temperature, breakup of the clusters and significant migration of point defects is known to occur.

Analysis of dislocation channeling in irradiated and deformed rhenium shows that most of the channels are parallel to $\{10\bar{1}0\}$ prism planes but some are parallel to basal planes. The twinning plane most frequently observed is $\{11\bar{2}1\}$ with $\{1102\}$ and $\{11\bar{2}2\}$ also observed.

Samples of JM and MRC molybdenum as well as rhenium have been prepared for elevated temperature irradiation in the swelling capsule. The molybdenum was annealed at 1850 C for 20 minutes and the rhenium annealed at 2000 C for 30 minutes. The samples were characterized by x-ray examination prior to encapsulation.

Postirradiation annealing to 400 C has not changed the irradiation-induced length increase in molybdenum crystals irradiated to 1×10^{20} nvt ($E > 1$ Mev) in the as-grown condition. As reported earlier, the length increase in these crystals was much less than that observed in crystals annealed before irradiation. These results are compatible with the hypothesis that the as-grown crystal substructure provides

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sinks for vacancies, lowering their concentration and reducing the magnitude of effects attributable to these defects.

Precision lattice parameter measurements on crystals irradiated to 1×10^{20} nvt have revealed an as-yet-unexplained inconsistency. Measurements with molybdenum K α radiation yield a lattice parameter value significantly lower than do the same measurements with copper K β or K α radiation. However, on annealed unirradiated crystals the use of these two wave lengths produces identical values. The discrepancy in measured values must be a result of the greater penetration of the harder molybdenum K α radiation (about 15μ compared to 2μ for copper K β). If a new surface is prepared by electropolishing more deeply, the same results are obtained, namely, a higher lattice parameter nearer the surface. In order to establish the nature of this phenomenon, which may involve the short-range migration of vacancies and their annihilation at the surface, further experiments are in progress with specimens having different exposures and post-irradiation annealing treatments.

The deformation behavior of molybdenum single crystals, which were prepared by triple-pass electron beam zone melting, is being studied. The crystals were seeded to yield a $\langle 149 \rangle$ axial orientation and tensile specimens were prepared by electrolytic machining. After machining, the specimens were annealed for 16 hours at 1300 C. Strain-rate change tests have been conducted at 77, 150, 196, 273, and 323 K, and it is found that the activation volume decreases with increasing strain, in opposition to the observations made on polycrystalline molybdenum. The values of activation volume, between 1 and $10 b^3$, strongly suggest the Peierls mechanism as the rate controlling process. Because of the tendency of molybdenum to deform on multiple slip systems, necking occurs readily at portions of specimens with minor deviations in cross-sectional area. The resulting decrease in uniform elongation has limited the present tests to approximately 15% resolved shear strain at best. Alternate methods for testing these 1/8-inch diameter crystals in pure shear, the fracture plane does not rotate into an orientation of high resolved normal stress, and extensive deformation by slip is possible. It is hoped that a technique will evolve which will result in satisfactory ductility of irradiated crystals at the lower temperatures. Five to 10% shear strain is the goal upon extension of testing to 4.2 K.

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.

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The effect of applied uniaxial tensile and compressive stresses on the $\alpha \rightarrow \beta$ transformation in plutonium was investigated to provide indirect information on the nature of this transformation. The results are characteristic of a martensitic transformation. Applied compressive stresses, up to 7000 psi, raise the $\alpha \rightarrow \beta$ starting temperature less than 0.1 C/1000 psi at a heating rate of 2 C/hr, whereas applied tensile stresses lower the $\alpha \rightarrow \beta$ starting temperature 1 C/1000 psi at a heating rate of 2 C/hr. This effect can be explained on the basis that the $\alpha \rightarrow \beta$ transformation is martensitic, but it cannot be explained on the basis that the transformation is diffusional. The normal component of tensile stresses and the shear components of tensile and compressive stresses favor the $\alpha \rightarrow \beta$ transformation in plutonium, but the normal component of compressive stresses oppose the $\alpha \rightarrow \beta$ transformation. This is in keeping with the hypothesis that the $\alpha \rightarrow \beta$ transformation is a martensitic transformation.

The investigation of the crystallographic differences between β_α and β_γ plutonium has been pursued further. It has not, as yet, been possible to make positive identification of the lines which appear in both diffraction patterns. This prevents, of course, the type of crystallographic analysis which would lead to information relative to whether the differences noted are the result of a basic change in the crystal lattice form or to an atomic rearrangement within the same lattice structure. There is no question now but that the two types of beta are temperature dependent and are not solely a result of the differences in parent phase, transformation processes, and stresses. It is possible, but, at this stage of the investigation, highly uncertain, that two distinct phases are involved. It is becoming increasingly evident, however, that as the plutonium purity reaches high levels the crystallographic differences between the two types of beta, as evidenced by x-ray diffraction, become more distinct and the temperature ranges of stability for each become more sharply defined.

E. USAEC/AECL COOPERATIVE PROGRAM

1. In-Reactor Creep of Zr-2.5 wt% Nb

The stress of the 300 C, in-reactor creep test on heat treated Zr-2.5 wt% Nb alloy was changed from 36,500 psi to 45,000 psi after 1620 hours of test time. Throughout the course of the test at 36,500 psi, the minimum rates during reactor operation were unresolvable. However, the rate appeared to increase during reactor outages. Creep rates were higher just after reactor startup than just before reactor shutdown.

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Work on the high temperature in-reactor creep capsule is progressing according to schedule. A heater test assembly is almost completed, and long-term heater tests will begin soon.

2. Microscopy Studies

A chemical polishing technique for thinning Zr-2.5 Nb alloys has been found successful. Thin film observations of age-hardened material reveal a duplex structure consisting of untransformed alpha-grains and a highly twinned alpha region which has been transformed from the beta upon quenching. This is in agreement with optical microscopic observations.

Particles in the untransformed alpha-grains, which were originally thought to be niobium precipitates are now believed to be hydrides which resulted from the polishing. Analysis of the phase diagram reveals that the amount of niobium in the untransformed alpha is very small and little, if any, precipitation of niobium would be expected during aging. Most of the precipitation will occur in the alpha region transformed from the beta. This has not been definitely verified experimentally.

3. Zr-Nb Pressure Tubing

One additional milled slot type crack propagation test was completed. An attempt was made to fail a 60% CW test piece at 300 C with a 1/2-inch long milled slot. The web of the milled slot failed at 70,000 psi hoop stress. Upon repressurizing to 300 C, the seal plug failed. In all tests completed to date, there has been little or no crack propagation at room temperature or above. Since the Zr-2.5 Nb alloy does not appear to be susceptible to brittle failure in the 30% and 60% cold worked condition, no further tests of this type will be made.

The uniaxial tensile tests have been completed and the analysis of test data begun. Preliminary analyses show that this material has a yield of about 90,000 psi at room temperature in the 30% CW condition.

Installation of the instrumentation for the 350 C stress rupture testing loop has begun. This was delayed because one of the pressure transmitters had to be returned to the manufacturer who had supplied the wrong range.

4. Fog-Cooled Tests

Experiments were performed to determine the boiling burnout conditions for an electrically heated 19-rod bundle test section with fog-cooled inlet conditions. The test section consisted of 0.612-inch OD Inconel

tubes, 76 inches long, arranged in concentric rings of 6 and 12 rods about a central rod. The wall cross sections of these tubes were such that the test section would produce a radial heat flux distribution in which the inner 7 rods had heat fluxes 80.5% of that on the outer 12 rods. The spacing between adjacent rods in the bundle was 0.040 inch and was maintained by means of ceramic warts at 6-inch intervals. The bundle was placed in a flow housing 3.25-inch ID with a radial spacing of 0.060 inch. The flow housing was, in turn, placed in a stainless steel pipe which contained the pressure of the coolant. The electrical connector at the inlet of the test section was constructed so that the electrical path was through the side of the pressure tube. In this manner, coolant flow disturbances due to the presence of the electrical connectors were minimized.

The test section was operated with both subcooled water and fog (wet steam) at the test section inlet. For the fog-cooled runs, a special inlet coolant mixing device, designed and constructed by AECL, was used. This mixing device received a slightly subcooled water stream and a nearly dry steam stream, and mixed them to provide the desired wet steam at the test section inlet. The device was provided with two sets of internal components. With one set, the water and steam were well mixed, and a homogeneous water-steam mixture was delivered to the test section. With the other set, the water was concentrated at the center of the tube causing the coolant between the test section and the flow housing to be primarily steam. The test section was operated with both sets of internal parts during the fog-cooled experiments.

To supply the subcooled water and nearly dry steam required by the mixing device, the flow to the test section was split into two streams. One stream went directly to the mixer. The other stream went through an electrically heated steam generator, where it was heated to 100% steam, and then to the mixing device.

A summary of the test conditions run is shown in the table below. A total of 76 burnout determinations was made.

<u>Condition</u>	<u>Inlet Configuration</u>	<u>Pressure Tube Wall</u>	<u>Inlet</u>	<u>Pressure psia</u>
			<u>Coolant Condition</u>	
I	Homogenizer	Smooth	Subcooled	1000
II	Homogenizer	Smooth	Subcooled	1200
III	Homogenizer	Rough	Subcooled	1000
IV	Homogenizer	Rough	Fog	1000
V	Concentrator	Rough	Fog	1000
VI	Concentrator	Smooth	Fog	1000
VII	Homogenizer	Smooth	Fog	1000

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Analysis of the data is not completed; however, some general preliminary conclusions have been formulated.

- a. The use of rougheners on the pressure-tube wall increased the boiling burnout heat flux. The increase ranged from about 10 to 50%, depending upon coolant conditions. The increase tended to be greater with higher flow rates. The rod surface temperature excursions upon the onset of boiling burnout tended to be significantly lower when the rougheners were used.
- b. The use of the concentrator for the fog-cooled runs had no discernible effect on the boiling burnout heat fluxes. The high concentration of liquid water on the bundle heat transfer surfaces at the test section inlet was apparently dissipated by the time the coolant had traversed the 6.3-ft to the outlet end of the test section where boiling burnout occurred.
- c. Operation at 1200 psia decreased the boiling burnout heat fluxes by approximately 10%.
- d. The boiling burnout heat fluxes obtained with this test section showed a weaker dependence on enthalpy than did data from a similar test section, but with 0.050-inch spacing between rods and 0.100-inch spacing between rods and pressure tube wall. The boiling burnout heat fluxes obtained with the present test section were lower at the lower outlet enthalpies but were about the same at the higher outlet enthalpies.
- e. Boiling burnout occurred on the center rod when the outlet enthalpy was low. It occurred on any one, and usually on several, of the inner 7 rods for the higher outlet enthalpies.

The test section was intact and completely operable when the experimentation was completed. Upon disassembly of the bundle, no evidence of any damage due to the high temperatures experienced during testing was found.

F. CUSTOMER WORK

1. Radiometallurgy Laboratory

The results of routine examinations and measurements are or will be reported as part of the sponsoring research and development programs.

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N-Reactor Rupture Examination. Three N-Reactor ruptures were examined during the month. The third, a driver element, was of particular interest to the Laboratory because of the extraordinary amount of decay heat involved. The problem was resolved by quickly sectioning off the areas of interest, thereby providing a greater heat-transfer surface. Even so, one small section caught fire while it was being photographed after it had been out of water only 20 minutes.

I-131 Release. Daily releases of I-131 to the atmosphere exceeded the prescribed limits for 300 Area facilities, particularly during the cutting and examination of short-term cooled N-Reactor and PRTR elements. The activated charcoal filter in the cell exhaust system was replaced, giving some improvement. However, modification of the system seems necessary to provide a long-term solution. A sample program was initiated to provide the necessary data for a complete analysis of the system needed before deciding on necessary modification.

High-Temperature Tensile Testing. The first successful tensile tests were completed at 850 C using grips and extensometer clamps designed to be used at temperatures up to 1650 C. The samples were flat control specimens of 304 and 348 stainless steel.

A new Instron console for recording stress-strain data at H-Cell has been received and put into service making available the Instron control console for room-temperature testing at I-Cell.

Stereo Zoom Microscope. Installation of the stereo zoom microscope and the remote positioner in F-Cell was completed.

E-Cell Metallography Facilities. Fabrication of a remote hand grinder for large samples (1-1/2, 2, and 2-3/8-inch dia.) was completed and functional tests were initiated. Four removable polishing pans were fabricated and installed on space orbital grinder for functional testing.

Project CAH-136, Service Addition - 327 Building. The air duct interference on the outside of the building was corrected and erection of structural steel was initiated.

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.

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A sampling of pressure-control bellows used in N-Reactor was submitted to determine the cause of their failure. Mechanical failure of some of these beryllium-copper bellows was found to be due largely to a fatigue mechanism which may have been accelerated by corrosion. Evidence of corrosion was found in the form of round-bottomed pits which extended as much as 20% through the wall thickness of the bellows. Information available was insufficient to determine the cause of this corrosion. The information necessary to evaluate the corrosion has been passed on to N-Reactor engineering personnel for their study and further evaluation.

During the report month 363 samples were processed. 619 electron micrographs and a total of 893 optical micrographs and macrographs were taken, 3040 negatives printed, and 11,421 prints processed.

3. High Temperature Lattice Test Reactor (HTLTR)

An 11-day, 1000 C operating period on the HTLTR mockup was completed. Gas contaminant concentrations peaked at 4000, 560, and 4800 ppm by volume CO, CO₂, and H₂O, respectively, after approximately 3 days of operation at 1000 C. These concentrations then showed a general decrease and were 2300, 260, and 3100 ppm CO, CO₂, and H₂O, respectively, at the end of the 1000 C operating period. Water concentration at the blower discharge reached a maximum of 880 ppm at a core temperature of approximately 800 C, and then decreased to 320 ppm at the end of the 1000 C operating period. During the first 6 days of operation at 1000 C, the water concentration in the gas leaving the cleanup towers averaged about 180 ppm due to water loading of the towers. At the end of the 6-day period the gas was directed through the alternate pair of towers resulting in a drop in the exit gas water concentration to an average of about 25 ppm.

During the operating period approximately two gallons of water were removed in the molecular sieve cleanup towers.

Temperature of the core was maintained during the operating period by automatic adjustment of the heating element power using one of the heating element thermocouples as the primary sensor. This mode of automatic control proved satisfactory with core temperatures varying less than ± 5 C about a mean in the 1000 C range. One of the 12 Inconel sheathed chromel-alumel thermocouples failed during heatup. The remaining thermocouples performed satisfactorily.

No malfunction of the heating elements was noted during the entire operating period. These four elements appeared to be in good condition with only minor attack at a few locations. All four exhibited some attack at the protruding, bus connection ends.

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Safety and Control Rods. The safety rod assembly, which was re-installed in the mockup, operated satisfactorily for 14 lift and scram cycles during the recent (third) run. During the 15th attempt to pull the rod, it dropped back into the core from about 85 inches out, apparently due to clutch slippage. From the fully inserted position, it was not possible to pull the rod. It was decided to continue the test run with the rod fully inserted rather than terminate the run prematurely. The third run was terminated on schedule, and the safety rod housing was opened so that mechanism operation could be observed. The mechanism could not lift the rod without manual assistance. The mechanism was then removed from the housing and tested on the bench. Against a spring scale the mechanism pulled about 75 pounds before the clutch slipped. Elapsed time between removal from the dry nitrogen system to testing on the bench was about 1-1/2 hours. The mechanism was then tested in a vertical position, mounted similar to the way it is in the mockup. It operated satisfactorily with the load of 55 pounds. There is no explanation for this phenomenon, but the difference may be due to the different atmospheres. A larger clutch is now being installed in the mechanism.

The control rod assembly also was installed in the mockup for the third run. It operated satisfactorily (20 cycles) until the core was at about 700 C. At that time some resistance to motion was observed in the last 1/10-inch of travel, as had been observed in the previous run. After the previous run the scram latch had been modified so the rod would scram under this additional load and the latch functioned satisfactorily. The next day with the core at about 800 C the rod was operated again. Again, resistance to motion was observed at the end of travel (perhaps slightly sooner than the previous day), but since it had functioned properly the day before, it was decided to run in to the limit switch. Before reaching the limit switch, there was a solid thump, shaking the actuator assembly. It was assumed that the outer rod shell had broken and this was verified by subsequent operation of the assembly. Operation of the horizontal rod was terminated for the balance of the third test run. At the conclusion of the run, the control rod was removed from the mockup. The shell was broken where it joins the mounting flange. Probable cause of the break was insufficient clearance between the TD nickel inner rod and inner sleeve pieces which caused the sleeves to seize on the rod, forcing the rod to expand toward the core rather than out toward the expansion spring. After the rod expanded, it contacted the plug in the end of the outer shell. Longitudinal loading of the outer shell caused it to break at its weakest point. There is additional evidence of differential expansion interference between the rod and inner sleeves since some of the inner sleeves show longitudinal cracks. The clearance problem has been resolved on the detail design drawings and will be incorporated in the assembly prior to the fourth run.

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Environmental Tests on HTLTR Materials. Exposure tests on pure B_4C in nitrogen plus 300 ppm H_2O at 1100 C continued. These tests, which will provide an estimate of the reactivity of the B_4C control material with the reactor atmosphere, are approximately 80% complete.

4. EBWR Fuel Elements

Irradiation Testing of EBWR Prototypic Fuel Rods. Five capsules with short lengths of vibrationally-compacted UO_2 -1.5 wt% PuO_2 fuel rods are still in the MTR and 27 capsules have been discharged. Maximum burnups attained with the impacted fuel are 5.0×10^{20} fissions/cm³ (17,700 Mwd/ton of fuel) and 5.3×10^{20} fissions/cm³ (18,000 Mwd/ton of fuel), respectively.

The 21-rod bundle (Fuel Element No. 6501) containing full-size EBWR fuel rods, currently under irradiation in the PRTR core, reached a burnup of 0.52×10^{20} fissions/cm³ (1850 Mwd/ton of fuel). Removal and replacement of one of the rods will be undertaken during the present PRTR extended outage.

The three-rod cluster tests continue to indicate satisfactory in-reactor performance. The clusters contain a total of 36 full-size (1.08 cm diameter by 148 cm long) EBWR rods.

5. Other Customer Work

Hydrogen Furnace. A safety review was conducted of a high pressure hydrogen furnace fabricated offsite. Inspection showed that the vessel had yielded at the top closure during pressure-testing at the vendor's plant. The equipment was returned for repair and control circuits are being designed to ensure adequate operating safety.

IPD - PCTR - Al-Pu Alloy and Thoria Fuels. Sixty-one fuel elements were involved in the project: 16 contained thoria and the remainder plutonium-aluminum alloy. The fuel elements are approximately 1-1/2 inches in diameter and vary in length from 7 inches to 35 inches. Eight have 0.005-inch thick end caps on one end of each fuel element. Approximately 7500 grams of plutonium and 197 pounds of ThO_2 were used in fabricating all fuel elements. All fuel elements were clad in aluminum. Pu-Al fuel elements are of I & E geometry; thoria fuel elements are solid cylinders.

Of primary interest in the fabrication of these fuel elements was the development encountered in welding, decontamination, and length control. It was found that length control and decontamination problems were best resolved by relieving the ends of the cladding tubes to receive the end caps. Considerable welding development work was performed, but

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the two most interesting considerations were the attachment of the 0.005-inch thick end caps and the use of 0.100-inch thick end caps on thorium fuel elements in direct contact with the fuel during welding. A number of approaches were considered for the foil end caps, but the only technique which could finally be applied successfully was an aluminum soldering technique.

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. The samples are tubular and made by coextrusion of core and cladding. In order to reduce scrap generation, increase the utilization of material, and permit more rapid fabrication, a die pressing technique was developed to preshape the plutonium alloys prior to extrusion.

Gold-aluminum alloy (stand-in for Pu-Al) cores were formed by the die pressing technique and coextruded. The core configuration of the extruded tubes was acceptable. The Pu-240 was received. Fabrication of the samples has begun.

Phoenix Fuel Wafer Production: Approximately 1000 Phoenix fuel wafers for PCTR experiments were produced from Al-20% low-exposure Pu alloy. Tooling and equipment needed to fabricate the required 9000 wafers were obtained and development of spiral machining techniques was completed.

Casting, extruding, and "spiral machining" steps are proceeding satisfactorily, although difficulties have arisen in cutting and coining the wafers to finished dimensions and quality standards.

G. NON-AEC RESEARCH & DEVELOPMENT

1. Technetium Alloys

Examination of the microstructure and determination of the phases present and their lattice parameters and microhardness was completed for a series of as-arc-melted W-Tc alloys containing up to 60 at.% Tc.

The structure of the alloys through 40 at.% Tc is single phase solid solution with decreasing lattice parameter as shown in the following table. Alloys with 20 at.% Tc and greater were severely cored and dendritic in appearance due to the rapid freezing of the melt button. Comparison of these lattice parameter values with those reported elsewhere indicate that the arc melt alloys were essentially quenched from approximately 1800 C, and that in this nonequilibrium condition,

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Lattice Parameters for Phases in the W-Tc System

<u>Composition at.% Tc</u>	<u>Phase</u>	<u>Lattice Parameters (°A)</u>	<u>Hardness DPH*</u>
Tungsten	Alpha Solid Solution (body centered cubic)	$a_0 = 3.1649 \pm 0.0001$	363
2.5	"	" 3.1635 "	318
3.5	"	" 3.1628 "	334
5.0	"	" "	376
10.0	"	" 3.1553 ± 0.0002	445
20.0	"	" 3.147 ± 0.002	528
30.0	"	" 3.134 "	518
40.0	"	" 3.126 "	495
50.0	Alpha Plus	" 3.117 "	470(matrix) 960(eutectic)
	Sigma (Tetragonal)	Not determined	530**
60.0	Alpha Plus	$a_0 = 3.117 \pm 0.002$	--
	Sigma (Tetragonal)	$a_0 = 9.520 \pm 0.001$ $c_0 = 5.003 \pm 0.001$ $c/a = 0.525$	1145**

* 1 Kg load 136° Diamond.

** 5 Kg load 136° Diamond (average value over two phases).

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the tungsten solid solution contains about 48 at.% Tc. Both the 50 and 60 at.% Tc alloys were two-phase in microstructure, and as indicated in the table, contain the b.c.c. alpha solid solution and the tetragonal sigma phase. Interpretation of these structures at the present time is that the 50 at.% Tc alloy is alpha plus eutectic (alpha plus sigma) and that the 60 at.% Tc alloy is primary sigma plus eutectic.

Additional evidence for this interpretation and determination of the composition of the sigma phase present is being sought by electron beam microprobe analysis of these two alloys.

The microhardness data, also shown in the table, indicates an initial hardness decrease with a minimum at 2.5 at.% Tc followed by solid solution hardening. A decrease in hardness was observed in the 30 and 40 at.% Tc alloys and in the matrix (alpha) of the 50 at.% alloy that is presumably related to the observed increase in ability of the alpha structure to deform by twinning. The second phase (eutectic) in the 50 at.% alloy is considerably harder and is brittle, being cracked by microhardness indents. The 60 at.% alloy is also extremely hard, with cracking evident in the matrix (eutectic) around hardness indents but not in the primary sigma phase. No twinning deformation was observed in the 60 at.% alloy.

Tungsten-technetium alloys have required special polishing techniques and etches to properly reveal the true microstructure. The as-cast buttons were especially vulnerable to surface defects such as twinning and smeared metal induced by the polishing processes. The polishing procedure as finally adopted for the series of alloys submitted for study was as follows. The samples were ground successively on 120, 240, 320, 400, and 600 grit silicon carbide papers with water as a coolant on an automet polisher. A rough polish with 6 micron diamond on microcloth was sometimes given the samples, again using the automet. Overnight polishing on a Syntron vibratory polisher followed using metcloth, Linde A abrasive and water. Final polishing was accomplished on microcloth with Linde A and a 1:1 mixture of sodium dichromate and chromic acid (each of 2% strength) for about two hours. The worked metal still present was removed by swabbing the samples with a solution of 45 parts by volume nitric acid, 45 parts water, and 10 parts hydrofluoric acid. Etching of the samples was done as follows. For alloys up to 40 at.% technetium a mixture of 10 g potassium ferricyanide, 10 g potassium hydroxide, and 100 ml of water was most effective. For 50 and 60 at.% alloys electrolytic etching with 10% oxalic acid proved to be better.

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2. Aluminum Honeycomb Extrusion

A 1.070-inch diameter, 0.035-inch wall Al tube containing a bonded 89 cell honeycomb structure has been successfully extruded. The web of the honeycomb is approximately 0.010-inch thick and the cells are randomly oriented and shaped.

The billet was an evacuated four-inch diameter, 0.100-inch wall Al can containing 89 Al clad Mg rods. Preheat temperature was 260 C and extrusion ratio was 14 to 1. The surface of the extrusion was excellent. Some distortion of cell size was caused on one side of the extrusion by either shift or nonuniform billet upset. The magnesium sacrificial material was readily removed by dissolution in concentrated nitric acid.

One attempt to roll the rod at 300 C into a flat failed due to the rod bursting diametrically during the second pass. Apparently neither the Al-Al nor Al-Mg bonds are strong enough to withstand the rolling stresses since both broke.

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