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Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

August 1964

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

August 1964

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FOREWORD

The Reactor Development Program Progress Report, issued monthly, is intended to be a means of reporting those items of significant technical progress which have occurred in both the specific reactor projects and the general engineering research and development programs. The report is organized in a way which, it is hoped, gives the clearest, most logical over-all view of progress. The budget classification is followed only in broad outline, and no attempt is made to report separately on each sub-activity number. Further, since the intent is to report only items of significant progress, not all activities are reported each month. In order to issue this report as soon as possible after the end of the month editorial work must necessarily be limited. Also, since this is an informal progress report, the results and data presented should be understood to be preliminary and subject to change unless otherwise stated.

The issuance of these reports is not intended to constitute publication in any sense of the word. Final results either will be submitted for publication in regular professional journals or will be published in the form of ANL topical reports.

The last six reports issued
in this series are:

February 1964	ANL-6860
March 1964	ANL-6880
April 1964	ANL-6885
May 1964	ANL-6904
June 1964	ANL-6912
July 1964	ANL-6923

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I. BOILING WATER REACTORS

A. BORAX-V

1. Operations

Power operation with the BORAX-V peripheral superheater core PSH-1 was terminated on August 14 after producing a total integrated power of 266 MWd. The final two weeks of operation were conducted with a prepared defective superheater fuel sample.

When the defective fuel sample was installed, seven boron-stainless steel poison rods were removed, reducing the number of poison rods to 33; and some of the remaining poison rods were relocated to positions shown in Figure 1. In addition, two 9.9-w/o enriched boiling fuel thermocouple rods were installed in place of 4.95-w/o rods. The fueled oscillator in the reflector, which had a broken drive shaft connection, was removed. This revised loading was designated core PSH-1B.

The reactivity gain of core PSH-1B (core PSH-1 had 40 poison rods) was about 0.5% at 120°F. This value does not account for the oscillator orientation which was unknown during the last critical experiments on core PSH-1 because of the broken drive shaft. The reactivity worth of the oscillator at room temperature had been determined to be only +0.009% or -0.015% with fuel section or poison section, respectively, facing the core.

A power of 22.4 MWt was attained with core PSH-1B by increasing power rapidly to minimize xenon buildup. Steam was vented directly to atmosphere, the feedwater temperature was 82°F, the reactor water level was 11 ft 10 in., and the bypass demineralizer (DM-1) flow was 40 gpm. This is the maximum power attained from any of the BORAX-V cores.

At equilibrium xenon conditions, the maximum power was 12.3 MWt with steam going to the turbo-condenser system. In this case, the feedwater temperature was 100°F, the reactor water level 11 ft 10 in., and DM-1 flow was 36 gpm.

During the two weeks of operation with the prepared defective superheater fuel, radiation levels throughout the plant remained low and operators were not restricted in their normal work of reactor and steam power plant operation. Radiation levels are given in Table I.

The stack monitor used to sample the air ejector exhaust system effluent has two sections: a gas and a particulate monitor. At equilibrium xenon conditions of 12.3 MWt, $6.6-7.2 \times 10^3$ cpm/ft³ for the gas monitor and $2.2-7.0 \times 10^3$ cpm/ft³ for the particulate monitor were observed. The stack effluent consisted of 6 to 10 cfm of air ejector exhaust flow and 120 to 140 cfm of dilution air flow. The gas-monitor readings with no defective fuel and with

the superheater fuel defect were similar, but values higher by a factor of 2 to 10 were observed during operation with the 10 defective boiling fuel rods. The particulate monitor gave the highest readings during the operation with the superheater fuel defect. Air monitoring 100 to 500 ft downwind of the air ejector exhaust stack and the atmospheric steam vents throughout the defective fuel tests indicated less than 1% MPC for unknown emitters.

Figure 1. Peripheral Superheater Core PSH-IB of BORAX-V

Table I. Plant Radiation Levels (mR/hr) during Defective Superheater Fuel Tests with Core PSH-1B

	Power Level (MWt)			
	12.3	16.5	18.9	22.4 ^{††}
Main Steam Line Reactor Building*	95-100	150	150	200
Main Steam Line Turbine Building**	40-50	75	90	-
Condenser Hotwell	40-45	70	110	-
Air Ejector Exhaust Charcoal Filter	130-150	190	210	-
Reactor Water Line to Bypass Demineralizer DM-1	60-85	80	75	-
Condensate Return Line [†]	12-15	18	20	-

*Monitor located on the main steam pipe. The pipe line distance from the reactor is about 42 ft.

**Monitor located on the main steam pipe. The pipe line distance from the reactor is about 300 ft.

[†]Monitor located on the condensate pipe. The pipe line distance from the condenser hotwell is about 300 ft.

^{††}Steam vented to atmosphere.

During the two-week operating period, miniature ion chamber traverses were made at 15 MWt, and experiments were conducted to determine the effect of reactor water level on reactor power.

2. Experiments

a. Defective Superheater Fuel Experiments. A sample of defective superheater fuel, a 1- x 1- x 0.020-in. piece of fully loaded peripheral superheater fuel plate, was prepared. The Type 304L stainless steel-clad cermet plate contained 31.8 w/o of highly enriched UO_2 dispersed in a matrix of Type 304B stainless steel. The cladding was removed from one side of the sample by selectively removing the stainless steel with aqua regia, thus exposing (confirmed by microscopic examination) the imbedded UO_2 particles, which remained essentially undisturbed. An 0.018-in.-dia stainless steel spacer wire was electron beam welded to each side of the two edges of the sample parallel to the steam flow path. As shown in Figure 2, a support strip and rod with a bayonet latch permitted the sample to be installed in the central steam, 0.060-in.-thick, coolant channel of a fresh second-pass superheater fuel assembly in the location shown in Figure 1. The sample was located 8-9 in. above the bottom of the fuel.

Figure 2. Defective Superheater Fuel Plate Sample and Superheater Assembly Used with Core PSH-1 of BORAX-V

Special steam-sampling lines were connected to the top of the riser on the superheater fuel assembly containing the defective sample and to the top of the riser of a normal second-pass assembly. Deposition samples in the form of 1/2-in.-dia rods were installed in the riser of the defective fuel assembly and at three locations in the steam line. Wafer-type deposition samples, 1-1/4-in. sq, were installed at the turbine exit.

Gross specific activities of steam and water samples from locations given in Table II were determined. As expected, the activity buildup in the reactor water was slowed down by the action of the reactor water demineralizer. With the demineralizer off, the activity (i.e., Na^{24} as noted in the past) builds up quite rapidly. Activities in the steam system from the defective superheater fuel sample were of the same order of magnitude as those from the ten defective boiler rods (see Progress Report for July 1964, ANL-6923, Table II, p. 3).

Table II. Gross Specific Gamma Activity (c/m/ml) Obtained with Defective Superheater (SH) Fuel Tests with Core PSH-1B

Sample Date and Time	Samples Taken from							Demineralizer Condensate S-11	Power Level	Demineralizer DM-1 Flow (gpm)
	Reactor Water S-4	Saturated Steam S-14	Defected SH Assembly S-16	Normal SH Assembly S-17	Superheated Steam S-13A	Steam to Turbine S-12	Condensate S-7			
Aug. 4										
1320	39,800	2,300	17,500	1,500	4,100	2,900	950	30	6.2	
1458	32,700	1,968	18,800	2,300	6,500	4,900	2,900	175	10.0	35
1540	71,700	2,500	21,600	2,900	6,200	4,600	2,700	180	15.0	38
1624	112,000	1,700	18,000	2,300	6,700	5,600	4,500	330	20.0	39
Aug. 5										
0905	113,000	2,400	17,000	2,200	6,100	3,800	2,100	210	13.0	38
1100	106,000	2,200	20,400	2,700	5,300	3,000	2,300	210	13.0	38
1253	110,000	5,700	20,000	2,300	10,400	4,100	1,800	145	13.0	38
1353	109,000	2,200	19,500	2,400	3,600	4,200	2,200	270	12.6	37
1534	111,000	2,000	20,300	2,400	5,900	3,700	1,300	350	12.2	37
Aug. 6										
0947	118,000	2,400	20,500	2,900	5,300	4,300	1,400	510	12.2	36
1300	125,000	2,500	15,600	2,500	5,900	4,100	2,700	460	12.4	36
1450	111,000	3,800	14,000	2,400	3,500	3,700	2,500	400	12.2	36
Aug. 7										
0935	252,000	2,400	8,100	2,400	5,800	3,500	2,400	466	4.5	10
Aug. 10										
1535	130,000	4,300	82,000	4,800	6,800		Steam to Atmosphere		22.0	40
1625	178,000	0	45,000	6,200	7,800		Steam to Atmosphere		4.0	0
1740	347,000	3,000	30,000	5,600	8,400		Steam to Atmosphere		15.0	0
1845	466,000	4,900	21,000	3,300	7,900	5,300	2,800	700	15.6	0
2000	251,000	8,500	25,000	3,200	7,600	6,500	1,700	600	15.0	39
Aug. 12										
0930	126,000	1,500	22,000	2,900	7,600	6,300	1,500	200	12.2	36
1545	127,000	7,200	25,600	3,700	8,100	4,947	-	-	13.5	39
Aug. 13										
1447	88,600	1,300	86,000	3,000	10,000	7,200	6,200	220	13.5	36
1610	111,000	1,500	87,000	3,100	7,100	-	3,600	260	16.4	37
Aug. 14										
0910	124,000	7,600	36,000	3,100	8,600	3,400	3,100	310	14.0	39
1118	119,000	3,500	22,500	2,800	4,000	3,700	2,500	460	13.6	39
1315	125,000	4,900	27,000	5,600	5,100	3,900	2,800	180	7.7	38
1540	110,000	-	37,000	3,400	7,100	5,500	4,000	440	13.0	38

Comparison of the gross counts obtained from condensed steam which has passed through the normal superheater fuel assembly and from the reactor water gives an idea of the practical decontamination factor characteristic of BORAX-V for approximately normal conditions (i.e., before defect tests, since the boiling section seemed to clean up rapidly after the defective boiling rod tests). These values, which include activity contributions from all sources, are shown in Table III.

Table III. Gross Decontamination Factors Determined in Defective Superheater Fuel Tests with Core PSH-1B

Superheated steam at exit to normal superheater assembly, 10 ft from fuel	45.5
Superheated steam at exit to defective superheater assembly, 10 ft from fuel	4.7
Saturated steam in steam dome	42.0
Steam line, 30 ft from reactor	18.0
Steam in line, 65 ft from reactor	45.0

Isotopes identified in the steam from the defective superheat assembly were I^{131} , Sr^{91} , Ba^{140} , and La^{140} . No Mo^{99} was found. The presence of these isotopes indicates also the presence of the gases Kr and Xe.

The I^{131} activities in Table IV were determined for all sampling points except the condensate. The iodine levels were low even in the steam from the fuel assembly containing the defective sample. Several unexplainable results are shown, such as: the highest I^{131} concentration was observed during a low-power run; more I^{131} was noted in the steam than in the reactor water initially; iodine levels were higher from the normal superheater assembly than from the main steam line.

Measurements made with samples of gas taken at equal volume and pressure from the air ejector system both before and after the charcoal filter indicated that the charcoal filter reduced the gas activity by a factor of 2.8.

The decay of the gross activity deposited in the main steam line was followed after the last shutdown. The major isotopes have not yet been identified, but the half-life of the gross activity has been determined. The half-life can be approximated as follows:

<u>Time after Shutdown</u>	<u>$T_{1/2}$</u>
0 hr - 30 min	7 min
1/2 hr - 34 hr	6.2 hr
34 hr - 70 hr	23 hr
Over 70 hr	84 hr

Table IV. I^{131} Activity (d/m/ml) Obtained in Defective Superheater
Fuel Tests with Core PSH-1B of BORAX-V

Sample Date and Time	Samples Taken from						Power Level	DM-1 Flow (gpm)
	Reactor Water S-4	Saturated Steam S-14	Defected SH Assembly S-16	Normal SH Assembly S-17	Superheated Steam S-13A	Steam to Turbine S-12		
<u>Aug. 4</u>								
1320	94	280	50	84	5	85	6.2	
1458	69	416	86	95	12	25	10.0	35
1624	57	79	44	58	9	10	20.0	39
<u>Aug. 5</u>								
0905	23	<5	110	20	7	12	13.0	38
1100	40	<5	41	<5	<5	11	13.0	38
1253	31	<5	33	<5	18	<5	13.0	38
1353	23	30	35	<5	22	<5	12.6	37
1534	19	6	23	7	<5	<5	12.2	37
<u>Aug. 6</u>								
0947	13	<5	83	<5	9	<5	12.2	36
1300	11	7	40	<5	5	<5	12.4	36
1450	11	5	29	<5	<5	8	12.2	36
<u>Aug. 7</u>								
0935	24	20	299	<5	46	13	4.5	10
<u>Aug. 10</u>								
1535	-	<5	119	26	22	14	22.0	40
1740	19	<5	162	35	13	17	15.6	0
1845	27	<5	65	13	14	15	15.6	0
2000	15	<5	41	16	9	8	15.0	39

Although gross radiation fields did not change greatly from the defective boiler to the defective superheater tests, deposition did show a significant change. The specific activities of deposition samples in the steam system were more than a decade higher from the defective superheater than from the defective boiler. The results from the deposition samples in the turbine exit showed that carbon steel acquired more activity by deposition by at least a factor of 2 than any other material in the steam system. The deposits on the samples all gave the same gamma spectrum, consisting primarily of peak energies from the isotopes Ba^{140} , La^{140} , and Co^{58} .

One-inch slugs cut from each deposition sample rod and counted gave results shown in Table V. The wafers in the turbine exit were also counted. These are arbitrary counts, but comparative, because the same geometry, count time, elapsed time (as near as possible), and surface area were used. All rod counts were made approximately 137 hr after the terminal shutdown. The counting geometry was slightly different for the square wafers. The wafer samples were counted approximately 69 hr after shutdown.

The deposition rod in the riser of the defective superheater assembly showed an abnormally high activity on the portion near the tip because

of neutron activation of the alloy. It was not possible to obtain a gross count of the first samples from this rod that was obviously related to the deposition. Deposition will have to be determined by radiochemical separations. Decreasing axial distribution of deposited activity along the steam line is obvious from Table V. Radial distribution was examined, but showed nothing definite. Deposition was approximately exponential for the first 65 ft only.

Table V. Steam-System Deposition Activity Obtained in Defective Superheater Fuel Tests with Core PSH-1B of BORAX-V

Rod in Defective Assembly Riser, Stainless Steel, 3-9 ft above Fuel		Rod in Steam Line Manifold, Stainless Steel, 24 ft**		Rod in Steam Line Test Section, Carbon Steel, 46 ft**		Rod in Steam Line Desuperheater Inlet, Carbon Steel, 69 ft**		Wafers at Turbine Exit, 300 ft**	
Sample Position, in. to Tip*	Count, C/cm ²	Sample Position, in. to Tip*	Count, C/cm ²	Sample Position, in. to Tip*	Count, C/cm ²	Sample Position, in. to Tip*	Count, C/cm ²	Sample Material	Count, C/cm ²
Tip	485,000	Tip	59,500	(***)		Tip	27,000	1-1/4 Cr-1/2 Mo	1516
1	446,000	1	59,400	1	32,000	1	26,600		
2	417,000	10	50,200	2	54,800	7	26,500	Type 304 SS	628
3	401,000	21	56,600	6	30,500	13	23,500		
4	374,000	33	59,800			19	32,500	Admiralty Brass	1261
5	347,000	45	89,200					Carbon Steel	2682
6	325,000								
7	306,000								
8	288,000								
9	274,000								
10	254,000								
16	185,000								
29	143,000								
32	162,000								
45	133,000								
58	91,000								

*These rods mounted axially. The tip is defined as the unmounted end.

**Pipe line distance from center of reactor.

***Test section rod was located transversely.

The flexible stainless steel hose that carried the steam from the defective superheater was destructively tested to determine whether or not more deposition occurred in the corrugated pipe than on a smooth surface. Activation of the stainless steel in this hose was negligible. Both gross specific activity and gamma spectrographic analysis indicated that the corrugations did not enhance deposition. The activity per unit area of flexible hose was almost the same as the activity per unit area of deposition sample from the top of the riser on the defective assembly. The deposition samples and the flexible hose had the same isotopes deposited on their surfaces.

b. Defective Boiler Fuel Experiments. Additional data from the deposition samples from the defective boiler fuel experiment (see Progress Report for July 1964, ANL-6923, pp. 2-6) have been analyzed.

Deposition data obtained with sample rods placed in the steam line were:

Stainless Steel Rod in Manifold, 24 ft	$7.3 \times 10^{-4} \mu\text{C}/\text{cm}^2 \text{Ba}^{140}$
Carbon Rod in Test Section, 46 ft	$2.42 \times 10^{-3} \mu\text{C}/\text{cm}^2 \text{Ba}^{140}$
Carbon Rod at Desuperheater Inlet, 69 ft	$4.16 \times 10^{-4} \mu\text{C}/\text{cm}^2 \text{Ba}^{140}$
Stainless Steel Rod in Manifold, 24 ft	$2.8 \times 10^{-4} \mu\text{C}/\text{cm}^2 \text{Co}^{58}$

These values are corrected to the time of shutdown, i.e., 1628 hr on July 20, 1964. Pipe line distance from the reactor is shown.

Figure 3. Power Level vs. Water Level at Equilibrium Xenon Conditions with Core PSH-1 of BORAX-V

Only small quantities of tritium were found in several steam samples at selected points in the system. The range of activity was 1.390 – $2.770 \times 10^{-3} \mu\text{C}/\text{ml}$. The highest value was obtained from the steam line on June 26 during a power level of 10 MW; the lowest level was found in the reactor water at a power level of 18.8 MW on July 20. The applicable limit of release is $3 \times 10^{-2} \mu\text{C}/\text{ml}$ based on a continuous release rate for 160 hr/week. As shown, the measured concentration is well below this limit.

c. Change in Reactor Power Level with Water Level. As the reactor vessel water level is increased, the reactor power level increases. The reactor was operated at maximum power (control rods fully withdrawn) until equilibrium xenon conditions were reached. Then the reactor water level was increased from 11 ft 0 in. to 12 ft 10 in., as measured by the level recorder LR-1, which provides a pressure-compensated differential-pressure measurement of water level. The power level computed from feedwater flow measurements increased from 11.8 to 13.7 MWt. Data are plotted in Figure 3. This increase is believed to be due to an increased natural-recirculation rate resulting from

the increased water level driving head and/or the reduced carryunder of steam with the recirculation water.

In a second run the water level was increased to 13 ft 2 in. to see whether or not the maximum power level due to this effect had been reached. Equilibrium xenon conditions could not be reached because of limited time. The highest power level (14 MWt) was reached at a water level of 12 ft 10 in. As the water level increased to 13 ft and 13 ft 2 in., the power level decreased. Reduced steam temperatures at the interpass plenum, superheater outlet, and steam line indicated that increased moisture was being carried into the superheater.

The acoustic water-level probe, mounted on the center nozzle of the reactor vessel top head, indicated that occasional splashes of steam-water foam were reaching a height about 11 in. above the level indicated by LR-1. The lowest superheater inlet is 4 in. above this height (14 ft 1 in.).

d. Boiling Fuel Rod Central Temperatures. Temperatures indicated by some of the thirteen boiling fuel rod thermocouples continued to show a general trend of increase with increasing burnup, although the trend seems to have decreased or reversed in two cases observed. Figure 4 shows the indication of a change in the pattern for one thermocouple in fuel rod 49, core position 56, during the last period of reactor operation. A similar thermocouple in fuel rod position 43 of the same fuel assembly showed a continued increasing temperature as a function of burnup (see Figure 5).

Figure 6 shows boiling fuel rod temperatures for rod position 43, core position 35 (which contains 9.95 w/o-enriched fuel), and rod position 43, core position 56 (loaded with 4.95 w/o-enriched fuel). One operating thermocouple of two installed in fuel rods 43 and 49 of core position 35 showed an early indication of a trend toward lower temperatures. The cumulative burnup values used refer to fuel burnup from the beginning of core PSH-1 operation, except for the values reported for fuel rod 43, core position 35. Burnup for this rod was zero when the balance of the core had reached an integrated power of 4,474 MWhr. Because of symmetry, the two groups of curves show the effect of fuel enrichment on operating temperature. The relatively early drop in temperature of rod 43 indicates that the fuel-rod conductance may be influenced more by time above a critical fuel temperature than by total burnup.

As is evidenced for Figures 4-6, the temperature drop is small compared with the general rise in temperature as a function of burnup. This would indicate two different mechanisms might be responsible for the changes noted.

Figure 4. Boiling Fuel Rod Central Temperature
vs. Reactor Thermal Power and Burnup
for Core PSH-1 of BORAX-V

Figure 5. Boiling Fuel Rod Central Temperature vs. Reactor Thermal Power and Burnup for Core PSH-1 of BORAX-V

Figure 6. Comparison of Boiling Fuel Rod Central Temperatures for 4.95% Enrichment vs. 9.95% Enrichment Obtained with Core PSH-1 of BORAX-V

e. Neutron Flux Distributions. Distributions of the axial neutron flux obtained from flux wire activation are shown in Figure 7 plotted on a relative count basis. In Figure 8 these are plotted on a point-to-average basis. The data were obtained in one core location at powers of 0.5, 5, 10, 15, and 20 MWt. The axial position of the peak flux in the boiler shifted 3 in. downward, that is, from 12 to about 9 in. above the base of the fuel, as the power increased from 0.5 to 20 MWt. The shift of location of the axial peak flux in the superheater region was only about half of that observed in the boiler region.

Figure 7. Axial Neutron Flux Distributions from Core PSH-1 of BORAX-V

Figure 8. Point-to-average Axial Neutron Flux Distributions from Core PSH-1 of BORAX-V

Measurements of neutron flux from the miniature ion chambers and flux wires were in very good agreement; axial flux shapes and cadmium ratios showed very similar characteristics for comparable conditions. It is interesting to note that both the flux wires and the miniature ion chambers produced cadmium ratios that were fairly insensitive to the void fraction (see Progress Report for July 1964, ANL-6923, pp. 4-11). Even at high power levels, there was very little change in the cadmium ratio versus axial height. The measurements did not show a decrease in the cadmium ratio in the region of increased voids, as was predicted.

3. Water Chemistry

The water chemistry parameters measured during the last two weeks remained essentially as noted for previous operations and are summarized in Table VI.

Table VI. Water Chemistry Parameters

	<u>Saturated Steam</u>	<u>Superheated Steam</u>	<u>Reactor Water</u>
pH	5.6-6	5-6	6-6.7
Resistivity, M Ω -cm	1.3	0.4-4.0	1-5
Chloride concentration, ppm	<0.04	<0.04	<0.04

II. LIQUID-METAL-COOLED REACTORS

A. General Fast Reactor Physics

1. ZPR-III

a. Assembly 45. Assembly 45 (see Progress Report for June 1964, ANL-6912, p. 16) is a multizoned reactor whose central region was specifically designed for studies of the fast-reactor Doppler effect in plutonium-uranium systems. The fuel mockup contains Pu^{239} and U^{238} in a ratio of 1:7, corresponding to the ratio to be expected in a large power-breeder reactor. The neutron energy spectrum has been intentionally softened by replacing some of the sodium cans with graphite to enhance the Doppler reactivity changes seen for U^{238} .

Radial traverses performed with U^{235} , Pu^{239} , and B^{10} , all sensitive to neutrons in the intervals of interest for Doppler measurement, have confirmed the calculated flatness of the low-energy neutron distribution.

Measurements of the Doppler coefficient of U^{238} are in progress. Initial studies have shown that controls of reactivity drift (caused by heating from the plutonium fuel) can yield a reactor stable to within 0.02 lh for several hours at a time, easily long enough to perform satisfactory measurements at any given Doppler temperature. Initial measurements at 500°K, chosen as the "cold" temperature to avoid crystal effects, showed a reactivity loss with respect to 300°K of approximately 0.48 lh; measurements to 1200°K, a conservative "hot" temperature for the present equipment, indicated a further reactivity loss for the samples used of approximately 0.9 lh.

The control of reactor drift possible with the present equipment promises to allow measurements with a precision exceeding that which was achieved on Assembly 43.¹

b. Doppler Measurements. A new series of experiments to measure the Doppler effect of natural uranium and fissile material necessitated the fabrication of new test samples. The first set of new samples were to be 1/2 in. in diameter by 6 in. long and assembled from pellets, 1/2 to 1 in. in length, fabricated from natural UO_2 , 7:1 UO_2 - PuO_2 , low Pu^{240} content PuO_2 , high-enrichment UO_2 , and 17% enriched UO_2 . A second set of new samples were to contain natural UO_2 fabricated in samples, 1 in. and 0.73 in. in diameter by 6 in. in length, to investigate rod-size effects on Doppler measurement.

The 1/2-in. natural UO_2 samples have been installed in ZPR-III.

¹G. J. Fischer, "Experimental Results for U^{238} Doppler Measurements in Fast Reactor Spectra," Proceedings of the Conference on Breeding, Economics and Safety in Large Fast Power Reactors, October 7-10, 1963, ANL-6792.

During the month, test elements containing the 1/2-in. 7:1 UO_2/PuO_2 samples were out-of-pile tested and shipped to Idaho.

The natural UO_2 samples for the rod size experiment were also assembled and tested.

The PuO_2 pellets were coated with water glass and fired to provide a glassy coating over the plutonium to reduce the contamination problem during handling procedures. The pellets are now being loaded into the sample cans.

Calculations of radial reaction rates were done for experiments performed in Assembly No. 45 with BF_3 , U^{235} , and Pu^{239} samples. The poor agreement obtained between theory and experiment was attributed to streaming in the counter hole and poor counter geometry. It was also found that the energy range of cross-section Set 224 does not extend low enough to describe the flux in the reflector region.

2. ZPR-VI

a. Axial Flux Traverses in Assembly No. 2. Axial flux traverses were made by the foil-activation technique in Assembly No. 2 of ZPR-VI, a 600-liter core containing uranium, sodium, and graphite with a length-to-diameter ratio of about 1:1.

The foil materials used were gold, copper, U^{235} , and U^{238} . The foils were round, of 1/2-in. (1.27-cm) diameter, and 5 mils (0.0127 cm) thick. Rows of eight foils lined up in axial direction were placed on top of drawers near the center position. The space between consecutive foils was 7.5 cm; the first foil was placed 2.5 cm from the interface of the reactor halves. As the depth of the core in each half was 45 cm, the first six foils of each series were located in the core and the last two in the blanket. All flux traverses were taken both with the reactor in its normal loading and with the central region (of 8-in. diameter) voided of sodium.

The gold-foil traverse in the core with the voided region was made with three sandwiches of three gold foils each which were put in the single-foil locations at 2.5 cm, 25 cm, and 47.5 cm from the interface of the halves. The sandwich technique was used to determine if a sufficient number of neutrons exist at energies as low as the gold resonances, the largest of which is at 5 eV. If this were the case, considerable self-shielding is to be expected due to the high-resonance cross sections, and a lower activation is expected of the center foil of the sandwich. Measurements showed that each foil in the sandwich was activated about equally, indicating that most of the activation took place at neutron energies higher than the resonance region. In the flux plot (Figure 9a), the average activation of the foils in the sandwiches is indicated by the triangular points.

Figure 9a. Traverses with Gold Foils

	Calc	Meas
With sodium in the test region	—	●
Without sodium in the test region	---	○
Without sodium in the test region	Avg of 3 foils in sandwich	△

Figure 9b. Traverses with Copper Foils

	Calc	Meas
With sodium in the test region	—	●
Without sodium in the test region	---	○

Figure 9c. Fission Traverses with U^{235}

	Calc	Meas
With sodium in the test region	—	●
Without sodium in the test region	---	○

Figure 9d. Fission Traverses with U^{238}

	Calc	Meas
With sodium in the test region	—	●
Without sodium in the test region	---	○

A comparison of the results of the measurements with flux traverses calculated by diffusion theory (RP-122 code using cross section set No. 199) are shown in Figure 9(a-c). In each figure the results are given for one foil material, with and without the sodium in the central region of the core. In order to clearly show the change in the flux shape which is caused in the center by the void, the curves were normalized in the outer region where the composition remains unchanged. Each theoretical curve and each series of experimental values were normalized so that the sum of the activations at distances of 32.5, 40.0, 47.5, and 55.0 cm from the interface was unity.

Except for some scatter, the measured points are generally in good agreement with the calculated values. The largest deviations are observed for the measurements with copper with the central region voided and with U^{238} with sodium in the central region. In the case of copper, this discrepancy was probably due to an excessively long time between irradiation and counting, resulting in low intensities and large statistical errors. (Error bars are given in the figures where the statistical error significantly exceeds the size of the points.) The deviations in the U^{238} traverse have not been fully explained.

3. ZPR-IX

a. Reflector Studies. Spatially dependent worth studies related to the properties of aluminum reflectors were reported for aluminum and carbon (see Progress Report for July 1964, ANL-6923, p. 17). Similar measurements for rhenium are reported in Table VII.

Table VII. Spatially Dependent Worths of Rhenium

Radial Position,* cm from Core Center	Weight, kg	Total Worth, lh		Specific Worth, lh/kg
		Relative to Aluminum	Relative to Void	
26.03	43.03	-131.5	-77.8	-1.81
30.16	43.03	-68.8	-34.8	-0.81
34.9	86.06	-66.6	-25.9	-0.24
41.4	71.69	-22.5	-10.2	-0.14
47.4	100.4	-8	~0	~0

*Sample position was weighted by the solid angle subtended to the core.

b. Fission Rate Traverses of U^{234} , U^{235} and U^{238} Detectors. Foils of U^{235} and U^{238} were irradiated in axial and radial positions in the core and reflector. Reaction rates of each material relative to its central reaction rate are shown in Figure 10. Also shown is the U^{238} to U^{235} fission ratio as determined from these foil traverses.

Figure 10. Radial and Axial Measured Reaction Rates and Fission Ratios of U²³⁵ and U²³⁸ Foils Relative to Measurements Made at the Center of the ZPR-IX Assembly No. 4

Solid-state detectors were also used for fission rate traverses. One traverse with a U²³⁵ detector compares well with the foil data (see Figure 10). A radial traverse with a U²³⁴ converter plate is also shown in Figure 10.

c. Central Reactivity Worths. From the viewpoint of reactor control, it is of interest to determine the degree to which hydrogen can be used to enhance the boron worth. Experimentally, this was determined by measuring the central worths of various homogeneous mixtures of powdered Lucite and amorphous boron. Specially fabricated sample cans of aluminum were filled with 63 cm³ of mixtures having known weight ratios of Lucite to boron. Because of the different densities of the materials (Lucite powder, ~0.4 g/cm³; solid Lucite, ~1.18 g/cm³; powdered boron, ~0.6 g/cm³; theoretical boron, ~2.34 g/cm³), the measured worths of the samples were normalized to unit weights. The measured points are shown as the circles in Figure 11. The worth per gram of boron was determined from the smoothed curve A, and is shown as curve B of Figure 11. It appears that a 65:35 w/o mixture of Lucite to boron has the maximum worth per gram of boron.

Figure 11

Central Reactivity Worths. Curve A shows measured reactivity worths of mixtures of boron and Lucite. Curve B shows those of boron as determined from Curve A.

A parametric study of the worth per gram of samples of different sizes was made with boron Lucite and boron powders. The results are shown in Figures 12 and 13. Unless there are significant amounts of self-shielding in these samples, there is no reason to expect them to depart from a constant value for all sample weights. Although errors are not shown, the scatter of points should be largest for the smallest samples. It is estimated that the value for the Lucite sample has a 25% experimental error and that of the $2\frac{1}{2}$ -g boron sample a 7% error. However, repeated measurements with the same samples indicated that the errors are about half these magnitudes. The curves drawn through the points are based on all the data obtained, only part of which is shown as the circles.

Figure 12. Measured Reactivity Worth of Lucite

Figure 13. Measured Reactivity Worth of Boron

Similar studies are currently being done with polyethylene.

d. Central Worth Coefficients. The central worth coefficients of a number of additional materials and central worth coefficients obtained in Assembly No. 4 are shown in Table VIII.

Table VIII. Central Worth Coefficients

Material	Sample Weight, g	Worth, lh/kg
Au	1183.6	-39.3
Zr	406	-1.07
ZrH ₄	107.01	+62.5

e. Central Gap Worth. Assembly No. 4 was the first of the ZPR-IX assemblies for which the central gap worth was measured. The results are shown in Figure 14. If the actual gap between the matrix halves were known when the tables are together, the gap worth could be used to correct the critical mass for the reactivity loss. In practice, a figure of 0.1% $\Delta k/k$ has been used for this correction, and it constitutes the largest uncertainty in the experimental critical mass. With the use of 430.5 lh = 1% $\Delta k/k$ for this core, the gap corresponding to 0.1% would have to be slightly less than 1/16 in. (0.159 cm), which is the estimated clearance between drawer fronts.

Figure 14
Effect of Gap between Reactor Halves of
ZPR-IX Assembly No. 4 on Reactivity

B. General Fast Reactor Development

1. Development of Jacket Materials

a. Vanadium and Vanadium Alloys. Jacket materials for the fuel elements of fast reactors must have suitable high-temperature properties, including strength and integrity, good thermal conductivity, and compatibility with the nuclear fuel and with liquid-metal coolants such as sodium. The jacket materials must also be amenable to fabrication into the desired shapes. Of the various potential jacketing materials that have been considered, vanadium-base alloys are among the more promising, and titanium is the element principally favored for use as an alloying element in the vanadium. Substantial additions of a third element have been tried, but any resulting increases in strength are usually obtained at the sacrifice of ductility.

Results to date indicate that, of the binary compositions, those with about 20 w/o of titanium (TV-20 alloy) are probably most suitable. Alloys with higher percentages of titanium have been made, but their hardness is greater and they are less easy to fabricate; alloys with lesser percentages of titanium are not so compatible with the fuel and are less resistant to corrosion in oxygen-bearing sodium. The commercial production of TV-20 is therefore being encouraged, and the fabrication development and properties of the alloy are being studied.

(i) Fabrication of Tubing. Continued fabrication of small-diameter (< 5 mm), thin-wall (< 0.5 mm) tubing of V-20 w/o Ti has led to methods that overcome many of the difficulties previously experienced (see Progress Report for April, ANL-6885, p. 13, for a summary of previous practice).

Currently, the extruded tube blanks are swaged, acid etched and vacuum annealed at 900°C for 1 hr. The annealed blanks are then drawn over a ductile core through aluminum-bronze dies. Wall thickness is adjusted at an intermediate stage by drawing over a beryllium-copper plug. Final sizing is also accomplished by plug drawing. Intermediate vacuum anneals are performed as required by the reduction schedule.

The use of nonferrous dies and plugs combined with a conventional draw lubricant has eliminated the galling problem. A much improved surface finish is obtained on both the outer and the inner surfaces of the tubes. A product controlled to closer tolerance also results from this fabrication technique.

(ii) Corrosion of V-20 w/o Ti in Sodium. A second experiment on the behavior of TV-20 in oxygen-bearing sodium (the results of the first experiment are given in Progress Report for July 1964, ANL-6923, p. 24) is in progress. Sodium peroxide was added in the amount needed to provide a concentration of oxygen in the sodium of 50 ppm. The peroxide, Na_2O_2 , rather than the oxide, Na_2O , was used to introduce the oxygen, because the peroxide is the only oxide of sodium that can be handled at room temperature in air of normal humidity without appreciable reaction with the moisture. The peroxide addition is, however, converted to Na_2O by reaction with a little of the free sodium. To obtain equilibrium solubility, the cold-leg temperature was brought to 232°C . After specimen-free loop operation for 16 days to allow the oxygen concentration to achieve equilibrium, the second run was started.

Present results show that weight gains are considerably higher for samples exposed to sodium at 50 ppm oxygen than for those exposed to sodium at 10 ppm oxygen. For an exposure time of 350 hr, the samples gained 0.85 mg/cm^2 and 0.46 mg/cm^2 for 50 and 10 ppm of oxygen, respectively.

(iii) Corrosion of Other Vanadium Alloys in Oxygen-contaminated Sodium. Although present results indicate that V-20 w/o Ti is reasonably resistant to the detrimental effects of oxygen-contaminated sodium on short-term corrosion tests, little information is available to allow comparison of the corrosion behavior of vanadium-titanium alloys in sodium with that of other vanadium alloys in the same environment. In connection with a program for the development of refractory-metal alloys for service in oxygen-contaminated sodium, the comparative effects on the corrosion behavior of various alloying additions in vanadium are being investigated.

One difficulty encountered in the pursuit of this program lies in obtaining reliable oxygen analyses of the sodium. In order to overcome this difficulty, a mercury amalgamation apparatus has been assembled and a vacuum sample-transfer apparatus is being built. It is believed that many of the present sources of error will be eliminated when these pieces of equipment are in operating condition.

Since the exact oxygen content (50-150 ppm) is uncertain, an experimental technique has meanwhile been developed for obtaining comparative data of some value. A piece of unalloyed vanadium is included as a comparison standard in each test, in which a set of samples is exposed to the molten sodium at 650°C . After exposure to the corrosive environment, the alloys are defilmed by immersion in cool $\text{KOH}\cdot\text{H}_2\text{O}_2$ solution and the weight loss per unit area is compared with that of the vanadium control. This permits a rough comparison between experiments involving differences in the respective oxygen contents.

A number of arc-melted vanadium-base 10 w/o binary alloys have been exposed. Titanium, molybdenum, and chromium additions provided the greatest increases in corrosion resistance. Aluminum, zirconium, tin, and yttrium additions appeared to be somewhat beneficial, whereas nickel, iron, and niobium additions caused increased attack by sodium at 650°C. Test durations were short (ca. 4-6 days). Since the samples were not homogeneous, the results must be considered useful only for preliminary screening. Alloys incorporating the beneficial elements are now being prepared in a levitation furnace in an effort to obtain castings that are more homogeneous.

C. General Fast Reactor Fuel Reprocessing Developments

1. Materials and Equipment Evaluation

A 10-in.-OD by 20-in.-high, welded tungsten crucible has withstood six thermal cycles between temperatures of 500 and 800°C without apparent effect. This crucible will be used for the next skull reclamation process demonstration run in the pilot plant.

Type 405 stainless steel is being evaluated as a container material for Cd-Mg-Zn-U/halide salt systems. Eight samples of Type 405 stainless steel were exposed to air at 650°C for 400 hr to test the oxidation resistance of this alloy. Weight gains of about 0.1 mg/cm² indicated excellent resistance to air oxidation. One additional sample was exposed at 650°C for 200 hr to a Cd-Mg-Zn-U/50 m/o MgCl₂-30 m/o NaCl-20 m/o NaCl-20 m/o KCl salt system at each of four positions in the melt: the metal phase, the metal-salt interface, the salt phase, and the vapor space above the melt. High corrosion rates were experienced by the samples that were exposed to the salt and vapor phases of this system, possibly because of hydrogen or hydrogen chloride liberated by the salt. This test will be repeated with salt of known low water content.

2. Advanced Processes

Cadmium-zinc-magnesium alloys are being evaluated as liquid metal solvents for the processing of uranium-plutonium fuel alloys. The stability of plutonium in Cd-Zn-Mg solutions toward various container materials is being investigated. Plutonium in a 70 a/o Cd-15 a/o Mg-15 a/o Zn solution in contact with 30 m/o NaCl-20 m/o KCl-50 m/o MgCl₂ at 600°C showed no significant interaction with Type 304 stainless steel (see Progress Report for June 1964, ANL-6912, p. 31) or with Type 405 stainless steel in 100-hr tests. In an identical preliminary test with 1020 mild steel, only 65% of the plutonium added was found in solution, thereby indicating that 1020 steel is not a desirable container material for this application. During the course of the solution-stability studies,

it was convenient to determine the distribution coefficients for plutonium in the Cd-Mg-Zn/molten chloride system. The distribution coefficient of plutonium $[K_d(\text{Pu}) = (\text{w/o Pu in salt})/(\text{w/o Pu in metal})]$ at 600°C was 4.2×10^{-2} throughout the 100-hr tests in all three container materials.

a. Preliminary Extraction Tests. Two preliminary high-temperature extraction runs have been made to develop techniques for separating plutonium from rare earth fission products by multistage extractions in molten metal-salt systems. The solute currently under investigation is uranium. In each run, 250 g of Cd-3 w/o Mg-0.5 w/o U alloy was poured into a static column of molten salt (130 g of 50 m/o MgCl_2 -30 m/o NaCl-20 m/o KCl) and packing which was contained in a quartz tube. In the first run (packing was 1/8-in.-dia glass spheres), performed at a temperature estimated to be 450 - 500°C , the salt contained 70% of the equilibrium uranium concentration after the molten metal was in contact with the salt for less than 5 min. In the second run (packing of 1/4 in. x 1/4-in. pieces of glass tubing), with the same metal-salt contacting time but a temperature of 630°C , the final uranium concentration in the salt was 90-100% of the equilibrium uranium concentration. Although some of the uranium transfer may have occurred between the metal collecting at the bottom of the column and the salt, the apparently high rate of uranium extraction provides encouragement that countercurrent extraction in a packed column would be successful.

3. Eddy Current Induction Probe

A new eddy current induction probe (see Progress Report for November 1963, ANL-6808, p. 25) for determining liquid levels of molten metals has been in service for two months without malfunction and without change in characteristics. The improved performance of the probe has resulted from the use of a grooved alumina form for supporting and separating the eddy current coils.

D. Sodium Coolant Chemistry

1. Analysis for Oxygen

A distillation analyzer has been modified by the addition of quartz windows to permit observation of the molten sodium in the sample cup during sampling and distillation. Results obtained with it have exceeded expectations. Leakage of sodium past the sampling valve and considerable variation in meniscus height have been observed. The height of the sodium meniscus above the edge of the sample cup appears to be a function of cleanliness (and wetting) of the outside edge of the cup. Contamination of this surface is believed to result from deposition of condensed distillate from neoprene gaskets and/or vacuum grease. No spattering of sodium, as originally hypothesized, has been observed during distillation.

2. Solubility of Carbon in Liquid Sodium

Investigation of a low-temperature, direct-oxidation method for the determination of carbon in sodium was continued. Complete recovery of carbon added as sodium carbonate was obtained, but carbon added as sucrose was only partially recovered, presumably because of loss as carbon monoxide. Use of a copper oxide furnace for conversion of the monoxide will be investigated.

E. EBR-II

1. Reactor Plant

a. Approach-to-power Experiments. The approach-to-power experiments (see Progress Report for July 1964, ANL-6923, p. 32) were continued. Reactor power was increased from 10 to 30 MWt, with an intermediate stop at 20 MW. Measurements of heat balance and transfer function were obtained at each power level and at various flows. Comparison with design is indicated in Table IX.

Table IX. Comparison with Design Power and Flow

<u>Power (MWt)</u>	<u>Design Power (%)</u>	<u>Design Flow (%)</u>
10	16	100, 74, and 36
20	32	100, 74, and 52
30	48	100 and 74

In a demonstration run the reactor operated continuously at 30 MWt during the period between 1330 August 13 and 0800 August 17 with an average electrical output from the turbine-generator of 7.5 MW. The demonstration run was terminated by a spurious signal which caused the reactor to scram.

The reactor was shut down on August 23 after a cumulative thermal power production of 343 MWd for the purpose of examining a core (2B1) and an inner blanket (6B2) subassembly as specified in the approach-to-power experiments. Three hundred forty-three MWd corresponds to approximately 0.1 a/o average burnup in the row two subassemblies. As of the shutdown date, August 23, the production of thermal and electrical energy was:

Thermal	343 MWd
Electrical	51.3 MWd
Reactor Critical	649 hr
Turbine-on Time	195.7 hr
Generator-on Line Time	174.9 hr

b. Power and Flow Coefficients. The bulk of the measurements for the transfer function are being made by use of the cross-correlation integrating device rather than the null-balance device (see Progress Report for July 1964, ANL-6923, p. 32). Although detailed analysis has not been completed, the data indicate that the reactor is stable under all conditions of measurement and that no instability is anticipated for the next incremental increase in power. Preliminary determination of the reactor power coefficient is in substantial agreement with that reported previously (ANL-6923, p. 33) for power up to 5 MW. The coefficient is approximately -2 lh/MW as measured with a constant reactor inlet temperature of 700°F .

Some stratification was observed in the primary tank during minimum-flow operation. At 10 MW and 36% flow, there was a maximum axial temperature gradient of 50°F over a vertical distance of about 15 ft.

Scram transients were performed at 20 and 30 MWt as planned. At 20 MW, two tests were carried out. The first involved reactor scram concurrent with loss of primary sodium flow with the auxiliary pump in operation. The second was conducted with the auxiliary pump de-energized. Preliminary data agree quite well with the calculations presented for these conditions in the EBR-II Hazards Summary Report (ANL-5719 Addendum). The tests involving loss of the auxiliary pump were terminated as planned when the primary sodium flow decreased to 0.5% because of the inability to measure in-core temperatures under zero-flow conditions. The transient test at 30 MW was conducted only once, with the auxiliary sodium pump deactivated. The conclusions agreed with those for the 20-MW test.

Routine analysis of the primary and secondary sodium indicated little change in the sodium in these systems. The plugging temperature of the sodium in the primary system has remained so low that it cannot be measured. In the secondary system, a pattern has established itself. The plugging temperature rises from 240°F to 300°F in about 5 days. After 2 days of operation of the temporary cold trap, the plugging temperature returns to 240°F .

c. Argon System. The primary cover gas of argon has shown a slow, but steady, increase in nitrogen content. Currently, gas-chromatograph analyses show about 1% nitrogen. Gamma-ray-spectrometer analyses of the cover gas at 30 MW reactor power show an A^{41} activity of $8.4 \times 10^{-4} \mu\text{C/ml}$, Xe^{135} activity of $5.9 \times 10^{-4} \mu\text{C/ml}$, and a trace of krypton activity.

In the secondary cover gas system, the gas chromatograph has shown a fairly constant nitrogen and hydrogen-helium level. Since the chromatograph cannot distinguish between hydrogen and helium, mass-spectrometer analyses were made. These indicate that the cover gas contains approximately 2200 ppm helium and 20 ppm hydrogen.

d. Sodium Pumps. Trouble was experienced in maintaining acceptable electrical winding temperatures in the secondary recirculating pump No. 2. The coolant channels were backflushed with clean water, removing considerable debris. The pump was returned to service with satisfactory cooling of the winding.

On August 2, following a reactor scram from 10 MW, an attempt was made to reverse pump voltage for reducing natural-convection flow. The voltage regulator apparently malfunctioned, putting full power to the pump in the reverse direction. Simultaneously, the main secondary EM flowmeter stopped functioning because of failure of an instrument connection probably caused by mechanical shock when the pump went into reverse. Recorded temperatures indicated a full flow reversal, and an increased level in the surge tank at the time also indicated the reversal. The pump was immediately shut down. A full investigation showed no apparent damage to the system. Since it had been recognized that the voltage regulator was unstable at very low voltages, the system will be modified accordingly; additional operating precautions are in effect until the modification is accomplished.

e. Temperature-measuring System. Analysis of heat-balance data taken during approach to power indicated a progressive failure of thermocouples located at the exits of the fuel-coolant channels. Leads to these thermocouples are divided into two groups of 13 each in the reactor cover-lifting columns. Sodium had leaked into one or more of the thermocouple tubes and progressively shorted out all 13 thermocouples in one group. The remaining 13 thermocouples are well distributed so that good surveillance of the outlet temperatures for core and blanket subassemblies is still possible. An attempt is being made to insert 1/16-in. stainless steel-sheathed thermocouples into the tubes after pulling out the existing thermocouples.

2. Power Plant

During operation, as reactor load was increased, heat was dissipated by bypassing steam via the bypass control valves to the turbine condenser. When a reactor load of 10 MW was reached, it was necessary to place all units of the cooling tower in service for the first time. Fans were started up and operated satisfactorily.

The steam line to the turbine-generator was blown to clear it of all foreign matter and debris. This was done by removing the bonnet from the turbine stop valve and installing a temporary connection to the outside of the Power Plant Building.

Upon completion of this operation, the turbine stop valve was re-assembled and the turbine was made ready for final checkout and startup.

A manufacturer's representative was present to supervise final checkout and startup of the turbine-generator. An air test was performed on the generator casing to assure tightness prior to startup and subsequent charging with hydrogen.

The 1250-to-150 lb pressure-reducing station was placed in service. With it in service, all Power Plant auxiliaries were supplied by steam from the reactor power plant, and the plant heating auxiliary boilers were placed in standby.

With the reactor at 20 MW and with air in the generator, the machine was brought to speed, synchronized, and loaded under manual control. All controls, including the initial pressure governor, were checked and adjusted where necessary. Overspeed trips were found to be satisfactory.

Following these checkouts, the generator casing was purged with carbon dioxide and charged to a pressure of $1/2$ lb hydrogen in preparation for the 30-MW demonstration run.

During the demonstration run, the turbine-generator was in operation under speed-governor control, with a small amount of steam bypassed to the condenser. After the demonstration run was completed, operation was continued to achieve the desired burnup of the fuel and for training purposes.

During the demonstration run and later, some difficulties were uncovered in the Power Plant. The most serious occurred after startup on August 17, when one of the bellows in the expansion joint of the steam-bypass line failed and allowed considerable air to leak into the condenser. The defective bellows was removed and the expansion joint temporarily modified to permit further operation. The expansion joint has been repaired. Studies have been initiated to determine the best approach to achieving a satisfactory and permanent correction to this difficulty.

The small control valve of the steam bypass performed poorly. Disassembly of this valve revealed extreme wear of the trim. The manufacturer has been contacted for recommendations to correct and avoid this situation in the future.

During the run following the demonstration run, the turbine's initial pressure governor was placed in service. Operation to date has been satisfactory. Until shutdown of the plant on August 23, operating crews of all shifts were trained in the startup and synchronizing of the turbine-generator and of the transfer of control from the speed governor to an initial pressure governor.

3. Fuel Cycle Facility

a. Development of Remote Controlled Methods and Equipment for Fuel Fabrication

(i) Fuel-decanning Equipment. Tests of operation of the decanning machine have indicated that chipping of the cutting-tool point with the originally specified tool material has occurred more frequently than is desirable. Recent tests of an alternative high-speed cobalt tool material have been more favorable: no tool chipping was observable after decanning 50 pins.

Resetting the tool closer to the fuel rod has improved feeding of the can scrap to the scrap cutter, and it has been found possible to eliminate the use of the scrap acceptor tube. This has eliminated the occurrence of scrap jams at this point.

The occurrence of bearing failures on the decanner when operated in the dry atmosphere of the Argon Cell prompted initiation of a program of tests of bearing materials and effects of atmosphere impurities. A group of samples is simultaneously tested, by rotating loaded polished steel cylinders against the samples, in a controlled-atmosphere box containing argon primarily. In tests of the effect of oxygen content on a selection of graphite samples in an atmosphere containing about 35 ppm water vapor and 1% nitrogen, wear was significantly greater at 10 ppm oxygen than over the range 50-1000 ppm oxygen. Tests of the effect of variations in the moisture content will be made next.

(ii) Fabrication of Additional Fuel Elements. In-cell production of acceptable fuel elements from the starting quantity of 36 kg of enriched fissium alloy was continued. The processing of an additional 129 acceptable fuel elements was completed through casting, demolding, inspection, assembly, welding, leak-testing, bonding, and bond-testing, to yield a total of 227 elements produced to date. About 57 more rods are in process, and a 7.2-kg ingot has been made available for a further injection casting run on this alloy. The latter was produced by melt-refining casting heels and pin scrap. The pouring yield in this operation was 92%.

(iii) In-cell Manipulators. It was reported (see Progress Report for July 1964, ANL-6923, p. 40) that during the development of a remote technique for manipulator carriage removal and replacement in the EBR-II Air Cell, a manipulator carriage assembly was accidentally dropped about 12 ft, from about bridge level onto a service stand located on the hatch cover of the transfer cell in the Air Cell floor. This accident resulted from the improper attachment of the crane hook to the lifting beam of the

manipulator carriage. This operation had to be carried out near the Air Cell roof, from outside the cell. Failure to achieve proper attachment was attributed to difficulties which were encountered in observing the hook-engagement operation (poor depth perception because of the 50-ft viewing distance involved and insufficient light). The manipulator carriage has been dismantled and inspected to assess the extent of the damage. The inspection showed that most of the major components can be re-used. The repair of the recoverable parts (major components and others) which were damaged is underway. Where repair is not practicable, replacement parts have been ordered.

b. Preparations for Operations

(i) Skull Reclamation Process. The process for the recovery of uranium from skull material remaining in a crucible after melt refining of EBR-II fuel is being studied on an engineering scale (charges of 1.5 kg uranium). The process steps are: (1) extraction (leaching) of noble metals from uranium oxide-halide flux into molten zinc, (2) reduction of uranium oxides to the metal with zinc-5% magnesium alloy and dissolution of uranium in the alloy, (3) precipitation of uranium-zinc intermetallic compound, (4) decomposition of the intermetallic compound and precipitation of uranium metal, (5) dissolution of the uranium metal and transfer of the solution from a primary vessel to a secondary vessel, and (6) retorting of the uranium product solution to concentrate the uranium.

Analytical results have been received for five engineering-scale runs (SKR-11 through SKR-15) performed earlier. In Runs SKR-11 and SKR-12, the waste flux was transferred from the primary to the secondary vessel along with the uranium product solution, instead of being transferred separately as in other runs, in order to produce a thick layer of flux on the alloy ingot which could be broken away easily and cleanly. When this modified step was used, cerium was back-extracted into the product solution and 1 and 7% uranium was lost in the flux. This modification is thus undesirable despite the good separation of flux from metal which would otherwise make this modification attractive. In Runs SKR-14 and SKR-15, the reaction times for the various steps of the process were decreased. Reaction times used in Runs SKR-13 through 15 are compared below:

	<u>SKR-13</u>	<u>SKR-14</u>	<u>SKR-15</u>
Noble Metal Leach, hr	4	1.5	2
Reduction to Metal, hr	2	1	1
Intermetallic Decomposition, hr	1	1	1
Dissolution of U Precipitate, hr	2	1	1

Reactions were successfully accomplished in the shorter reaction times, and there is evidence that further reductions in reaction times are possible. Total run times for Runs SKR-13 through SKR-15 were 26, 27, and 20 hr.

Data on the removals of ruthenium, molybdenum, cerium, and zirconium were obtained. Satisfactory ruthenium removals were obtained in all five runs. The molybdenum concentrations in the product solutions from the five runs represented 15, 23, 39, 41, and 44% of the molybdenum originally charged; these removals are satisfactory. Cerium in the product solutions from Runs SKR-13 through SKR-15 represented 5 to 13 percent of the cerium charged, as compared with previously reported values of 25 to 37 percent. Zirconium removals were somewhat low in all five runs. Evidence has been obtained in small-scale runs that some zirconium coprecipitates with the uranium-zinc intermetallic and is carried over to the product dissolution step.

Two additional engineering-scale runs have been completed. A problem of flux foaming was encountered in these runs. It is believed that this problem is associated with water of hydration present in the flux. Apparently, the water of hydration is not completely driven off when the flux is held under argon for several hours at 700°C during noble metal extraction. Methods are being examined whereby the water content of the flux and of the process system can be lowered.

The last step of the skull reclamation process is being demonstrated in engineering-scale (15 kg Zn-Mg) retorting runs that are being carried out by the Chemical Engineering Division at Argonne, Illinois. This step consists of the distillation of residual magnesium-zinc from uranium and the consolidation of the uranium. The demonstrations are being conducted in a modified melt refining furnace which is installed in a glovebox containing an argon-5 v/o nitrogen atmosphere. Three additional retorting runs have been carried out in which evaluation was continued of beryllia retorting crucibles produced by different fabricating techniques and manufacturers (see Progress Report for July 1964, ANL-6923, p. 40). In these runs, isopressed (~1/8-in.-thick wall) and thixotropically cast (~3/8-in.-thick wall) beryllia crucibles experimentally developed by the Coors Porcelain Company were tested.

In one run made with the isopressed crucible, the charge consisted of zinc-13.5 w/o magnesium. At the completion of the run, an examination of the crucible showed the presence of a great number of hairline cracks. In addition, a portion of the conical bottom of the crucible easily broke away.

Two runs were made with the thixotropically cast crucible for the recovery of uranium from two 5- to 6-kg charges of zinc-12 w/o magnesium-10 w/o uranium ingots (nominal composition). This crucible was also used in a previously reported retorting run (see ANL-6923, p. 40). Following each of the two current runs, 100 to 150 g of a metallic product was found in the graphite secondary crucible, which

indicated leakage of the melt through the beryllia crucible. The melt is believed to have leaked through cracks which were visible in the lower 2-3 in. of the beryllia crucible and which became more pronounced with each use of the crucible. Some of the cracks appeared to have penetrated the entire wall thickness. [A thixotropically cast beryllia crucible by the Brush Beryllium Company had performed satisfactorily for nine retorting runs (see ANL-6923, p. 40) before it failed because of handling.] Further work with Coors isopressed and thixotropically cast beryllia crucibles has been deferred. However, additional retorting runs with a new Brush Beryllium Company thixotropically cast beryllia crucible are being planned.

F. FARET

1. General

The principal activities for the current reporting period continued to be concerned with general Title II engineering at the laboratory and at the Architect-Engineer offices. Bechtel's efforts remain near schedule. Laboratory review of Bechtel's drawings is increasing significantly.

Considerable effort is being devoted to evaluation of design principles associated with containment in the immediate vicinity of the reactor vessel. Shielding and containment design, instrument thimble locations, and associated cooling problems continue to require major attention. The Bechtel reactor vessel design package has not been transmitted formally to the Laboratory; however, sufficient information has been received to permit preparation of the Laboratory's portion of specifications and drawings without further delays.

There has been an increase in the number of progress prints being received from Bechtel for review by the Laboratory. Major submittals recently received include electrical schematics, sodium piping drawings, argon system piping and instrumentation diagrams, process and service system piping and instrumentation diagrams, heating and ventilation drawings, piping layouts, and plant arrangement drawings.

Recent transmittals to Bechtel by the Laboratory, in addition to routine comments on drawings, specifications and design information, have included interbuilding coffin details, personnel air lock requirements, cell lighting data, and miscellaneous comments on various FARET ANL Data Transmittal Items.

Subsequent to the recent revision of design Package VII to accommodate early procurement and construction plans of United Engineers and Constructors, Inc., Bechtel has proposed that additional information be

included in Package VII. Their proposal would include specifications and drawings for the argon purification system, refrigeration equipment, radioactive liquid waste pumps, and various tanks associated with the waste system.

2. Safety Analysis

The Commissions' comments on the FARET Preliminary Safety Analysis Report have been reviewed and, as required, changes or additions have been made. The Report is in process of being published. The revised report will be transmitted to the Commission.

Work is continuing on programming an analytical model for a two-parallel coolant loop which will provide a better understanding of the operating characteristics of the reactor main and auxiliary coolant systems. These systems (loops) are mutually coupled in the reactor but independent otherwise. This program is required in order to properly evaluate system performance under various operating conditions, including scram, for both safety analysis and engineering design purposes.

3. Reactor Vessel

The reactor vessel procurement package will include the double-walled vessel, the core support cylinder including the reactor grid, the vessel conical extension piece, and the vessel cover. The design of the double-walled vessel with support is the responsibility of the Architect whereas the remainder is designed by the Laboratory. The total design package should be ready in early September.

A two step procurement procedure is planned which will consist of 1) prequalification of equipment vendors by a selection board and 2) firm price proposals by qualified vendors. The first step has been initiated and vendor responses are expected by the last of the month. The second step will begin in early September.

4. Cell Shielding Windows

Preliminary evaluation of the strength of the proposed glass slab of 6-in. thickness for the FARET cell window indicates that, under a maximum peak cell pressure of 30 psi, the deflection of the slab will be approximately 0.002 in. and the corresponding stress approximately 500 psi. A design stress of 800 psi used for commercial applications is well below the ultimate strength of 800 psi $\pm$ 20% for annealed glass.²

²Memorandum, J. F. Davis, Technical Conference with Pittsburgh Plate Glass Company, June 3, 1964

Little information is available regarding the impact strength of thick glass slabs. An attempt has been made to evaluate the damage to a slab under the impact of an object. If it is assumed that all the kinetic energy of an object is absorbed in locally crushing the glass, then

$$t = \frac{\partial V^2 W}{S_c g A},$$

where

t = depth of penetration, in.

V = velocity of object, ft/sec

W = weight of object, lb

S_c = crushing strength of annealed glass, 36,000 psi

g = gravitational constant, ft/sec²

A = average area affected within glass taken as twice the average cross-sectional area of the portion of the object penetrating the slab.³

Accordingly, the depth of penetration in a slab depends on:

- (i) the square of the velocity of the object;
- (ii) the ratio of the weight of the object to the average cross-sectional area of the penetrating object.

A procedure for testing the glass slabs as a structural member to indicate if they will satisfy the criteria of pressure, temperature and impact (see Progress Report for June 1964, ANL-6912, p. 49) has been developed as follows:

a. Hydraulic Pressure Test. The fixture for the hydraulic test has been designed. The available glass slabs (29 x 44 x 8½ in. thick) will be bolted down over the fixture with a Koroseal gasket between the optical face of the glass and the machined face of the bottom plate of the box. A heavy steel lid will be laid across the top and bolted to the sidewalls of the fixture to form a leak-tight enclosure. Either water or oil will be introduced and pressure will be built up with a hand hydraulic pump to about 150 psig. This pressure on an 8½-in.-thick slab of glass will be necessary to stress the glass to about the same value as a pressure of 60 psig would produce on a 6-in.-thick slab. This 60-psig pressure is twice the expected peak pressure of about 30 psig estimated for the case of a sodium-air reaction within the cell.

Micrometer dial gauges at two locations will be used to observe gasket compression when the glass is installed and the compression under

³Memorandum, A. H. Heineman, Destructive Potential in Case of BER Turbine Rotor Disintegration, March 4, 1955.

hydraulic pressure. Additional dial gauges, mounted to the bottom-plate opening, will measure the deflection of the glass slab.

The hydraulic test will be preceded and followed by Freon leak tests of the enclosure with glass and steel cover in place.

b. Free-fall Mechanical-drop Test. A free-fall mechanical-drop test will be performed with the hydraulic test fixture and a second framed glass similar to the first. The steel cover will be left off the fixture, and the second slab will be mounted on top of the first with suitable cushion gaskets between them. The fixture is designed so that when set on a level surface, the glass surfaces will be about 6° from horizontal. Such an angle should reject a dropped object so that the object will not fall back onto the glass a second time.

Initially, a steel ball of about 20 lb will be dropped from a height of about 20 ft. The depth of penetration will be compared with the results of theoretical analysis. A high-speed motion-picture record of the test will be made. The use of other instrumentation, such as strain gauges, is under consideration.

c. Thermal Shock Test. A two-slab assembly will also be subjected to thermal shock by sudden exposure of the outer glass to heat radiation from a vertical bank of infrared heaters, about 3 ft x 4 ft, spaced a couple of inches from the glass surface. After bringing the heaters to temperature, a fireproof insulating sheet will be removed from between the heaters and the glass. After a short time the heater power could be reduced to some fraction of the initial value and maintained for, say, 20 min, after which the power would be turned off, and the bank of heaters would be removed. The assembly would be allowed to cool and damage assessed. Following the heat exposure it is hoped to be able to remove the outer slab assembly, reinstall the steel cover, and conduct a Freon leak test on seal integrity of the inner slab.

5. Fuel Assembly Sodium Flow Test Facility

A pump similar to that in the large TREAT loop has been ordered for service in the 1200°F flow test facility. Several modifications in the design of the pump are needed for adaption to the service requirements of the flow test facility. The pump tube has been provided with eccentrically mounted transition sections to improve the ability to drain the loop. Bus bars of circular geometry have been designed in order to facilitate better sealing of the bus-bar penetrations through the gas-tight enclosure of the loop.

A tentative design for the freeze plug has been established. The structures required to support the pressure vessel, pump, expansion tank, and dump tank have been installed. The instrument racks, transformers, and associated equipment are available for installation and assembly. Initial facility operation should commence in December, 1964.

III. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. Mass-energy Relations in 6.0-MeV-neutron Fission of U^{235}

Two parameter measurements with solid-state detectors have been made on the fission of U^{235} by 6-MeV neutrons. Alternate measurements were made with the detector oriented at 0° and 100° to the neutron beam direction and at thermal energies. The preliminary results are given in Table X.

Table X. Preliminary Results Obtained from Two Parameter Measurements of U^{235} Fission Induced by Thermal and 6.0-MeV Neutrons

Neutron Energy: Detector Angle:	Thermal	6.0 MeV		
		0°	100°	All
Average Mass of Light Peak, $\bar{M}_L$, amu	96.4	98.1	96.7	97.6
Variance of Light Peak, σ_L^2 , amu ²	34.1	41.7	48.2	44.9
Average Mass of Heavy Peak, $\bar{M}_H$, amu	139.2	139.3	138.4	138.9
Variance of Heavy Peak, σ_H^2 , amu ²	33.3	45.4	43.2	44.7
Average Total Kinetic Energy $\bar{E}_T$, MeV	166.5	165.7	165.9	165.8
Total Kinetic Energy Corrected for Neutron Emission, $\bar{E}_{KT}$, MeV	168.3	168.1	168.3	168.2
Peak to Valley Ratio	~ 470	24.4 ± 2	22.7 ± 2	23.4 ± 1.4
Number of Fissions	132,123	19,420	13,908	33,327

The most prominent difference at the two energies is the greatly increased yield of symmetric fission at 6.0 MeV. Little of the neutron energy appears in the kinetic energy of the fragments (see values of $\bar{E}_{KT}$ in Table X). The average kinetic energy of the fragments in very asymmetric fission ($M_H > 150$) is appreciably higher in the 6.0-MeV measurements. This balances the increased yield of low-energy, symmetric fragments to give the same value of $\bar{E}_{KT}$ at both energies.

2. Total Neutron Cross Sections with an Automated Facility

An appreciable effort has been devoted to the automation of the accelerator and nuclear instrumentation in the Reactor Physics high voltage

laboratory to obtain increased economy, precision, and efficiency in the determination of fast-neutron cross sections. This goal has been achieved in some of the simpler types of measurements, particularly fast-neutron total cross sections. The system is so arranged that a CDC-160A digital computer controls all of the experimental operations, records all of the data, carries out on-line data analysis, produces the required cross sections, and then makes the executive decisions requisite for the next experimental step. This closed loop is schematically indicated in Figure 15.

Figure 15. The System Components and Their Relation to the Closed Loop

A particular application of the system to the determination of the fast-neutron total cross sections is indicated in the flow diagram of Figure 16. The sequence is as follows:

- a. The experiment is initiated at a given neutron energy.
- b. The computer calculates a Q -value to determine the accelerator beam energy necessary to produce the desired neutron from the $\text{Li}^7(p,n)\text{Be}^7$ reaction.
- c. The accelerator is then set to this energy by the computer.
- d. The bias for rejecting the undesired second neutron group from the source reaction is then calculated and set.
- e. The sample is removed from the beam of neutrons striking the detector.
- f. Data are taken into the computer from the monitor and from the detector.
- g. A continual test for statistical precision is carried out; when the specified criteria are satisfied, the sample is put into the neutron beam and the transmitted flux determined to the specified precision.

h. The cross section is then derived from the well-known relation

$$\sigma \sim \ln \left[\frac{\phi_{\text{trans}}}{\phi_{\text{incid}}} \right]$$

and recorded on a printed output.

i. The neutron energy is reduced by a desired amount and the sequence repeated, energy by energy, to a specified limit of lower energy.

Figure 16. The Logic Employed in Closed-loop Measurements of Total Neutron Cross Sections

A given cycle through the loop takes a minute or two. Representative total cross sections determined with the above automated system are shown in Figure 17. The curves for iron and aluminum display the typical resonance structure of light and intermediate nuclei at an incident neutron energy of ~1.0 MeV and are determined with resolutions of 1-3 keV. The curve for natural uranium is representative of the cross section of a heavy material near 1 MeV and is determined with a resolution of ~5.0 keV. Similar results have been obtained for a number of other nuclei and over an extended energy range.

3. Stripping Reactions

The nuclear reactions $\text{Be}^9(\text{d},\text{n})\text{B}^{10}$, $\text{Al}^{27}(\text{d},\text{n})\text{Si}^{28}$, and $\text{F}^{19}(\text{d},\text{n})\text{Ne}^{20}$ have been experimentally studied with deuterons

of energies from 2 to 7.0 MeV. The results are important because of the use of these reactions as neutron sources and because of the understanding of nuclear structure in light nuclei resulting from the measurements. The experimental procedure was based on fast-neutron time-of-flight techniques with the Reactor Physics Division intense, pulsed Van de Graaff and the Physics Division Tandem Van de Graaff Accelerator of higher energy. More than twenty neutron groups have been observed in the F^{19} and in the Al^{27} reactions, resulting in greatly improved knowledge of the structure of the product nuclei. Angular-distribution measurements now being made will define the angular momentum changes involved in the various processes.

Figure 17

Representative Total Cross Sections Obtained with Automated Closed-loop Operation

Detailed determinations of the angular distributions of ten neutron groups from the $\text{Be}^9(d,n)\text{B}^{10}$ reaction have been completed. The resolution is sufficient to resolve all neutron groups up to B^{10} excitation energies of 7.0 MeV. A typical experimental time-of-flight curve obtained at a deuteron bombardment energy of 3.2 MeV is shown in Figure 18. The resolution is approximately 15 keV. The peaks shown in the various curves obtained at different angles were integrated to obtain the angular distributions. A

Figure 18

Time-of-flight Distributions from $\text{Be}^9(d,n)\text{B}^{10}$. The four spectra were obtained simultaneously and represent increasing neutron sensitivity, top to bottom. The inset of neutron groups of progressively lower energy is thus clearly evident. The excitation energies of the various levels in B^{10} are indicated.

Figure 19. Angular Distribution of Neutrons from $\text{Be}^9(d,n)\text{B}^{10}$ Leading to the Ground State. The solid curves represent Butler calculations with a stripping radius of 5 Fermis.

thallium are complete, and the cross sections for the inelastic excitation of 6 excited states are shown in Figure 20. Less complete results for Re, Ta, and W indicate inelastic cross sections of 1.5-2.0 b at 1.0 MeV with values nearly as large extending down to incident neutron energies of several hundred keV. These large inelastic cross sections must lead to appreciable fast-neutron energy degradation in reactors employing these high-temperature materials.

5. High-conversion Critical Experiment

The microparameter measurements were completed on the core assembled during July, which had a stainless steel-clad central fuel zone and an atom ratio of 0.5 hydrogen atom per atom of U^{238} (H/28 ratio).

The U^{238} capture cadmium ratio measured in the 0.5 H/28 composition core was 1.035 ± 0.001 . The derived ratio of epi-to-subcadmium capture rates in U^{238} was 28.3 ± 0.5 . The measurement was made only by the

representative result is shown in Figure 19 where the angular distribution of neutrons leading to the ground state of B^{10} is given. The solid curve indicates the results of Butler calculations assuming the change of angular momentum, Δl , is equal to 1. The agreement between theory and experiment is excellent, confirming that this particular state is characterized by a positive parity and spin 3. The results shown in this figure are representative of those obtained for the other neutron groups.

4. Fast-neutron Scattering

The fast time-of-flight apparatus has been principally devoted during the past month to the determination of inelastic neutron scattering from the heavy elements Ta, W, Re, and Tl, the first three of which are of particular interest in high-temperature reactor applications. The measurements for

thermal activation (or substitution) method because there was insufficient room in the lattice to cover a fuel element with cadmium as required in making the direct measurements. The analyses of data pertinent to other microparameter measurements are in progress.

Figure 20. The Measured Inelastic Excitation Cross Sections of Thallium. The solid curves represent the results of optical model calculations.

Small systematic differences have been found in weight-normalized U^{238} capture rates, depending on the foil or pellet size, which were irradiated in the graphite thermal column of the Argonne Thermal Source Reactor (ATSR). These irradiations were made to standardize initial conversion ratio (ICR) measurements in the Hi-C cores. As a result of this effect, further investigations may require small adjustments in ICR values reported earlier.

Since a strong spontaneous fission source has recently been made available, an investigation to determine the capture-to-fission ratios in low-flux reactor systems has been resumed. The proposed method requires the determination of fission-neutron worth with a source of known strength and the core spectrum neutron worth by an absolute measurement of the capture rate by a manganese sample.⁴

Oscillation of a Cf^{252} source yielding $\sim 10^6$ spontaneous fission neutrons/sec was performed in a core having a 1.24-cm pitch square lattice and containing 948 aluminum-clad Hi-C fuel pins. As reported previously,⁵ a detectable response was obtained earlier by oscillation of a 6×10^4 -neutrons/sec source, but a 10^6 -neutrons/sec source strength appeared necessary to obtain reasonable precision. These new measurements demonstrated that the magnitude of the response to a source remains constant regardless of reactor flux level in contrast with fuel or poison oscillation, which produces a response proportional to flux level. The increased source strength now available appears adequate to determine the fission neutron importance function. Measurements with manganese and fissile samples are now underway.

⁴W. C. Redman, Pile Oscillator Determination of Capture-to-fission Ratio, TID-17738 (1962).

⁵Reactor Development Program Progress Report for September 1963, ANL-6784, p. 34.

B. Theoretical Reactor Physics

1. Matrix Representation of Detection Systems

The iterative method (see Progress Report for September 1963, ANL-6784, pp. 34-5) has been employed to unfold the resolution response of the lithium-sensitized solid-state neutron spectrometer. Since the resolution of this detector is approximately independent of neutron energy, measurements made at zero neutron energy (i.e., with thermal neutrons) define the response matrix completely. Table XI presents such a response matrix obtained for the $\text{Li}^6(n,\alpha)$ solid-state detector.

Table XI. Response Matrix for the Neutron Detector

Column No. Row No.	1	2	3	4	5	6	7	8	9	10
1	1.0000	0.8142	0.5948	0.3585	0.1843	0.1338	0.0871	0.0622	0.0467	0.0345
2	0.8244	1.0000	0.8142	0.5948	0.3585	0.1843	0.1338	0.0871	0.0622	0.0467
3	0.5083	0.8244	1.0000	0.8142	0.5948	0.3585	0.1843	0.1338	0.0871	0.0622
4	0.2310	0.5083	0.8244	1.0000	0.8142	0.5948	0.3585	0.1843	0.1338	0.0871
5	0.0579	0.2310	0.5083	0.8244	1.0000	0.8142	0.5948	0.3585	0.1843	0.1338
6	0.0209	0.0579	0.2310	0.5083	0.8244	1.0000	0.8142	0.5948	0.3585	0.1843
7	0.0054	0.0209	0.0579	0.2310	0.5083	0.8244	1.0000	0.8142	0.5948	0.3585
8	0.0010	0.0054	0.0209	0.0579	0.2310	0.5083	0.8244	1.0000	0.8142	0.5948
9	0.0019	0.0010	0.0054	0.0209	0.0579	0.2310	0.5083	0.8244	1.0000	0.8142
10	0.0024	0.0019	0.0010	0.0054	0.0209	0.0579	0.2310	0.5083	0.8244	1.0000

Figure 21 represents results of a typical experimental measurement of interest taken in the center of a fast critical reactor assembly. In order to employ the iterative method, the matrix representation

$$\underline{Y} = \underline{A}\underline{X} \quad (1)$$

($\underline{Y}$ includes the inherent experimental error) was symmetrized by multiplication on the left with $\tilde{A}$, the transpose of the response matrix A . One then treats the system

$$\underline{V} = \underline{B}\underline{X} \quad (2a)$$

with

$$\underline{B} = \tilde{A}\underline{A} \quad (2b)$$

and

$$\underline{V} = \tilde{A}\underline{Y}. \quad (2c)$$

In addition to the experimental data, or $\underline{Y}$ -vector, Figure 21 also displays the $\underline{V}$ -vector which results from the symmetrization process. The approximations obtained for 300, 360, and 420 iterations are depicted in Figure 22.

The peak which arises in the neighborhood of Channel No. 27 and Channel No. 28 corresponds to the resonance in the $\text{Li}^6(n,\alpha)$ reaction at 0.26 MeV. This peak, therefore, affords a convenient energy calibration for the detector.

Figure 21

The Li^6 Solid-state Detector Pulse-height Distribution or $\underline{Y}$ -vector and the Symmetrized Experimental Data or $\underline{V}$ -vector

Figure 22
The Iterative Approximations $\underline{X}(300)$, $\underline{X}(360)$, and $\underline{X}(420)$ of the Neutron Spectrum [weighted by the $\text{Li}^6(n,\alpha)$ cross section]

2. Effective Cross Sections for Reflectors by Use of ELMOE Code

In the calculation of the critical mass of a fast reactor assembly, the ELMOE code^{6,7} is often employed to obtain the effective group cross sections for transport and elastic transfer. Since ELMOE is strictly a

⁶H. H. Hummel and A. L. Rago, An Accurate Treatment of Resonance Scattering in Light Elements in Fast Reactors, Proc. of Seminar of Physics of Fast and Intermediate Reactors, SM-18/45 (1961).

⁷A. L. Rago and H. H. Hummel, ELMOE: An IBM-704 Program Treating Elastic Scattering Resonances in Fast Reactors, ANL-6805 (1964).

fundamental-mode-analysis code, it was originally believed that it was not applicable for use in either the outer regions of a reflected core or in a reflector region containing light-scattering materials, such as steel, sodium, and aluminum. However, the ELMOE code has been used for ZPR-III⁸ and other fast critical assemblies which were primarily U^{238} reflected. Now there is interest in using light-element reflectors, which introduce the problem of light-element scattering resonances and fine-group averagings into the calculations.

To obtain an indication of the sensitivity of critical mass to the choice of light-element group cross sections in a reflector, three sets of aluminum cross sections have been considered in studying the aluminum-reflected system of ZPR-IX Assembly No. 1 (see Progress Report for February 1964, ANL-6860, p. 10). The ELMOE code was used to generate two cross-section sets subject to certain simplifying assumptions on buckling and source spectra. The three cross-section sets used (see Table XII) in the comparison were:

- (a) the YOM aluminum cross sections;⁹
- (b) the ELMOE-determined aluminum cross sections in an applied fission spectrum source assuming zero buckling.
- (c) the ELMOE-determined aluminum cross sections in an applied constant source as a function of energy and assuming zero buckling.

Table XII. Comparisons of Group Cross Sections for Aluminum (barns)

Energy Group	Group Lower Energy, MeV	Cross Section Sets					
		Set a		Set b		Set c	
		Transport	Elastic Transfer	Transport	Elastic Transfer	Transport	Elastic Transfer
1	3.668	1.37	0.118	(1.37)*	(0.118)		
2	2.225	1.60	0.162	1.36	0.238	1.33	0.198
3	1.35	2.00	0.216	1.90	0.321	1.90	0.299
4	0.825	2.48	0.332	2.10	0.378	2.09	0.368
5	0.5	2.97	0.404	2.77	0.481	2.77	0.478
6	0.3	3.17	0.420	2.90	0.466	2.90	0.462
7	0.18	3.82	0.484	2.66	0.468	2.66	0.467
8	0.11	5.02	0.632	2.40	0.547	2.40	0.546
9	0.067	5.98	0.690	2.17	0.497	2.17	0.496
10	0.0407	2.78	0.308	1.97	0.323	1.97	0.323
11	0.025	6.34	0.721	0.639	0.202	0.638	0.202
12	0.015	0.75	0.081	0.711	0.106	0.711	0.106
13	0.0091	1.13	0.121	1.10	0.163	1.10	0.163
14	0.0055	1.51	0.161	1.49	0.184	1.49	0.184
15	0.0021	1.37	0.0507	1.37	0.105	1.37	0.105
16	0.0005	1.37	0	(1.37)	0		

*Parentheses indicate use of same value as in Set (a) for criticality comparison in cases of no ELMOE values obtained in these groups.

⁸Reactor Physics Constants, 2nd Edition, ANL-5800 (1963).

⁹S. Yiftah, D. Okrent, and P. A. Moldauer, Fast Reactor Cross Sections, Pergamon Press, New York (1960).

The ratio of critical core volumes (masses) found by using cross-section Set b relative to cross-section Set a in the reflector was calculated to be 1.17. The cross sections of Set c are so similar to those of Set b that no subsequent criticality calculations were made with Set c.

Current studies of reactors using light material reflectors show that the choice of an applied fission source together with the assumption of zero buckling leads to negligible error in reflector group transport cross sections. However, such is not the case for the group transfer cross sections. In a reflector region, the transport cross sections should be the more important parameter insofar as critical core mass is concerned. Therefore the calculated factor 1.17 found for the aluminum-reflected assembly should be a reasonable indication of the error which would arise if YOM aluminum cross sections are used.

3. Resonance Interference

The effect of moderator mass on the Doppler coefficient at 1000 eV has been estimated by a method based on Forti's recent paper.¹⁰ This effect is small for typical U^{238} resonances. For a heavy moderator the derivative of the resonance integral (of a homogeneous mixture) with respect to temperature is proportional to

$$\left(1 + \frac{1}{16} \frac{\sigma_m}{\sigma_p} \frac{\Gamma_p}{E_0} M\right),$$

where

σ_m = moderator scattering per heavy atom

$\sigma_p = \sigma_m + p_0 \sigma_{p0}$

Γ_p = practical width of the resonance

E_0 = resonance energy

p_0 = Forti's parameter

M = moderator-to-neutron mass.

For a typical fast reactor composition and average U^{238} parameters the correction factor is $(1 + 6.5 M)10^{-5}$ and is negligible.

4. Total Neutron Cross Sections

A study was made to determine whether the neglect of the finite resolution involved in transmission experiments causes appreciable error

¹⁰G. Forti, Evaluation of Resonance Integrals in Homogeneous and Heterogeneous Systems - An Intermediate Approximation, Nuc. Sci. and Eng., 19, 4 (1964).

in resonance parameters derived from fitting Breit-Wigner resonances to cross sections as a function of energy. In this study a CDC-160A code was written to calculate apparent "experimental" cross sections according to the equation

$$\sigma_{\text{app}}(E) = \frac{1}{n} \ln T(E),$$

with

$$T(E) = \int_{-\infty}^{\infty} e^{-n\sigma(E')} R(E' - E) dE'.$$

Here $\sigma_{\text{app}}(E)$ is the apparent cross section at energy E , $T(E)$ is the transmission, and $R(E' - E)$ is the resolution function, which was taken to be a Gaussian with width ΔE at half-height. Of course, if $R(E' - E)$ were equal to $\delta(E' - E)$, the apparent cross section would be the actual cross section.

The apparent cross sections from the 160A code were used as input for the 2092/RP least-squares-fit code on the IBM-704. Calculations were done for both $\ell = 0$ and $\ell = 1$ resonances at 750 keV for various resonance widths Γ and various resolution function widths ΔE . The option in the least-squares-fit code was used in which $g\Gamma n$ was varied in direct proportion to Γ . It was found that the error $\sigma\Gamma$ in Γ , produced by neglect of the finite resolution, was surprisingly small. For the rather extreme case that $\Delta E = \Gamma$, $\sigma\Gamma$ was only about 11% of Γ for $\ell = 0$ resonances and 4% of Γ for $\ell = 1$ resonances.

The reason for the small errors in Γ is that the use of the option with $g\Gamma n$ directly proportional to Γ implies that the calculated height of the resonance will be essentially correct even though the finite transmission may depress the apparent height considerably when ΔE is not small in comparison with Γ . Thus, the least-squares-fit code increases Γ somewhat to fit the apparent cross sections better in the wings of the resonance, but cannot improve the fit much near the peak. Therefore the sum of the squares of the deviations of the calculated fitted cross sections from the "experimental" values increases rapidly with increasing $\Delta E/\Gamma$, but $\sigma\Gamma$ increases much more slowly.

5. Interpretation of Rossi-alpha Data

A study has been made of the theory of Rossi-alpha measurement with a time analyzer that records only one event per sweep. This type of time analyzer is compatible for use with a multichannel analyzer which is used to store data when performing Rossi-alpha measurements. In the reduction of such data, one starts with the raw channel counts, C_i , and

obtains P_i , which is an estimate of the probability that a sweep proceeding as far as channel i will terminate with an event in channel i by applying the formula

$$P_i = \frac{C_i}{N_0 + \sum_{j=1}^n C_j}, \quad (1)$$

where n is the total number of channels, and N_0 is the number of sweeps during which no events at all are recorded. The quantity C_i is governed by binomial rather than Poisson statistics, but the estimate of standard deviation on P_i is not much different from the Poisson prediction for the usual case that P_i is small.

From one-group point-reactor kinetics, the probability of observing an event in the small time interval from t to $t + \Delta t$ is given as

$$p(t) \Delta t = (A + B e^{-\alpha t}) \Delta t, \quad (2)$$

provided that there has been an event at $t = 0$ irrespective of occurrences at intermediate times. From this, by using a derivation analogous to that used to obtain the standard Poisson distribution, it may be shown that

$$P_i = 1 - C_1 e^{-C_2 e^{-\alpha T_i}} \quad (3)$$

For the usual case with $C_2 \ll 1$, this reduces to the form

$$P_i = (1 - C_1) + C_1 C_2 e^{-\alpha T_i}, \quad (4)$$

which is commonly used to fit such data. It may be shown also that fixed or variable time delays between an initiating event and the actual commencement of a sweep do not affect the data reduction in practical cases.

For situations such as with reflected reactors, in which an additional decaying exponential term appears in Eq. (2), it may similarly be shown that

$$P_i = 1 - C_1 e^{-(C_{21} e^{-\alpha T_i} + C_{22} e^{-\alpha_2 T_i})}. \quad (5)$$

This is analogous to Eq. (3) and the same considerations apply.

C. High-temperature Materials Development

1. Ceramics

Among the fertile and fissile materials being studied or developed for reactor use at a temperature in the range from 750°C to 2000°C are

the ceramic compounds of uranium, of plutonium, and of thorium, and various mixtures of solid solutions of these compounds. Some of the binary compounds have melting temperatures of about 3000°C. Although most of the work on ceramic fuels has been on the oxides and the carbides, the sulfides, phosphides, and such combinations as UP-US, UC-US, PuS-US, and PuP-UP are receiving considerable attention. Eventually the arsenides, selenides, antimonides, and tellurides will be investigated.

Some of the more important considerations, besides melting point, are thermal, chemical and irradiative stability, fabricatability, composition control, the effects of impurities, compatibility with cladding materials, mechanical properties, volatility, thermal conductivity, and thermal expansion. No one of the binary compounds has all the desired characteristics in sufficient degree, but no one of the compounds is beyond the possibility of improvement through certain modifications and combinations of the compounds. Considerable emphasis is therefore being given to preparing these ceramics and studying their properties.

a. Measurements of Thermal Properties. Measurements of specific heats and thermal diffusivities of ceramic reactor materials at room temperature are being continued before putting the high-temperature equipment in operation. Graphite standards are run concurrently with the unknowns and data are obtained at various energy levels on specimens of various dimensions. This is to prevent variations in equipment and boundary conditions from affecting the results, which are shown in Table XIII.

Table XIII. Thermal Properties

Material	Theoretical Density, %	Thermal Diffusivity $\alpha(\text{cm}^2 \text{sec}^{-1})$	Specific Heat C_p ($\text{cal gm}^{-1} \text{C}^{-1}$)	Thermal Conductivity K_p ($\text{cal gm}^{-1} \text{cm}^{-1} \text{sec}^{-1}$)
Uranium				
Monophosphide	73.5	0.053	0.045	0.018
"	82.6	0.051	0.046	0.020
"	88.8	0.054	0.046	0.023
Uranium				
Monosulfide	96.4	0.040	0.045	0.019

No previous experimental specific heat values are available for uranium monophosphide. The specific heat of the compound calculated

from the sum of its constituents, uranium and phosphorus, is equal to $0.047 \text{ cal gm}^{-1} \text{ C}^{-1}$ at room temperature. Both the calculated and the experimental¹¹ value for uranium monosulfide is equal to $0.045 \text{ cal gm}^{-1} \text{ C}^{-1}$ at 25°C . As can be seen by comparing these values with the experimental ones of this report, the heat-pulse method is well-suited for the determination of the heat capacity of ceramics at room temperature. It is hoped that the reliability in the determination of this property can be extended to higher temperatures.

The values of the thermal conductivities of uranium monophosphide and monosulfide are considerably higher than that of uranium dioxide, reported previously, which was $0.014 \text{ cal gm}^{-1} \text{ cm}^{-1} \text{ sec}^{-1}$. The phosphide appears to be an appreciably better conductor of heat than the sulfide.

b. Anelasticity of Certain Compounds of Uranium. In conjunction with a program for the thorough study of the properties of uranium ceramics, a study has begun on the anelasticity¹² of compounds of uranium with some of the elements in Groups V and VI of the Periodic Table. The present objective is to measure the temperature dependence of the elastic moduli (and of related properties such as internal friction) of materials that show promise for use in high-temperature reactor applications.

(i) Apparatus for Dynamic Measurements of Elastic Modulus and Internal Friction. The apparatus for dynamic measurement of elastic modulus and internal friction has been assembled on a preliminary basis. A block diagram of the apparatus, as it now is, appears in Figure 23. A frequency counter, which will permit the determination of resonant frequency with more exactitude, will be added as soon as it is received.

Figure 23

Block Diagram of Apparatus for Measurement of Elastic Modulus and Internal Friction

¹¹ Westrum, E. F., Jr., and Gronvold, F., "Chemical Thermodynamics of the Actinide Element Chalcogenides," Thermodynamics of Nuclear Materials, IAEA (1962), pp. 3-37.

¹² Anelasticity: behavior such that no part of the stress-strain curve in the elastic region is single-valued. This behavior is caused by internal friction and other mechanisms that damp vibration.

The driver was a record-cutting head, Astatic X-26, with needle Astatic CL 92-3, and the pickup phonograph cartridge, Astatic L-12 with needle N4-38. The coupling wires were platinum, of 0.25-mm and 0.13-mm diameter. The larger diameter seems to be the better. Tests were performed at room temperature in order to determine the reliability of the apparatus. A single-crystal sapphire rod obtained from Linde Company was chosen for this determination. This sample, 15.24 cm long, was flame polished to a diameter of 0.381 cm. The orientation was C-axis $56^\circ \pm 5^\circ$ to rod axis. The flexural resonant frequencies, from which Young's modulus can be calculated, varied approximately from 1380 to 1410 cps, depending on the type of coupling between the transducers and sample. The internal friction, Q^{-1} , was determined from the decay of free vibrations of the sample by use of the relationship

$$Q^{-1} = \ln(A_0/A_1)/\pi f \Delta t,$$

where A_0 is the amplitude of the first free vibration occurring after a time Δt and f the resonant frequency. A characteristic pattern, obtained with the present setup, is shown in Figure 24. The values of Q^{-1} recorded were higher than 1×10^{-3} . Wachtman and Lam¹³ have determined the elastic modulus and internal friction of a sapphire rod of similar dimension. The orientation of this rod was: $X = 56^\circ$, $\lambda = 57^\circ$, where X is the angle between rod axis and projection of rod axis on the (0001) plane and λ is the angle between rod axis and the (1120) direction nearest the rod axis. They reported a flexural resonant frequency of 1381 cps, which agrees fairly closely with our determination.

Figure 24

Decay Pattern of a Sapphire Rod.
Vertical axis: 0.1 V/cm.
Horizontal axis: 50 msec/cm.

¹³Wachtman, J. B., Jr., and Lam, D. G., Jr., Young's Modulus of Various Refractory Materials as a Function of Temperature, J. Am. Ceram. Soc., 42 (5), 254-60 (1959).

The same authors obtained a peak width, Δf , about 0.3 cps at room temperature. From this value, the resonant frequency, and the expression

$$Q^{-1} = \frac{0.5773 \times \Delta f}{f},$$

can be calculated an internal friction of 1.25×10^{-4} , which is lower than our value. It should be pointed out that they calculated the internal friction by a different method than that used here, and their test was performed under vacuum, which eliminates the effect of air damping.

Another source of error might be due to the position of the suspension wires. Wachtman and Tefft¹⁴ have stated that an error of about a factor of 20 could result from neglect of the effects of suspension and air damping.

The results of our tests indicate that the apparatus gives fair results for Young's modulus, and that this accuracy will be increased by the addition of the frequency counter. In the case of the internal friction, the technique should be improved in order to eliminate as much as possible the effect of external damping. In tests with a steel bar of rectangular cross section (0.603 cm x 0.402 cm) and 15.2 cm long, it was not possible, owing to the weight of the specimen (33.1 gm), to detect the resonant frequency when suspending the sample directly from the coupling wires. This inconvenience was solved by resting the bar on supports located at the nodes and placing the transducers so that there was practically no stress transmitted to them through the coupling wires. The flexural resonant frequency of this sample was approximately 1040 cps. Use of this value in the equation

$$E = 0.94645 C m f^2 / B,$$

where

E = Young's modulus (dynes/cm²);

m = mass of the test piece (gm);

f = resonant frequency;

B = dimension of the cross section perpendicular to the direction of vibration;

C = shape factor¹⁵

¹⁴Wachtman, J. B., Jr., and Tefft, W. E., Effect of Suspension Position on Apparent Values of Internal Friction Determined by Forster's Method, Rev. Sci. Inst., **29**, 517-20 (1958).

¹⁵The shape factor was obtained from Tables for the Computation of the Shear Modulus and Young's Modulus of Elasticity from the Resonant Frequencies of Rectangular Prisms compiled by D. P. H. Hasselman.

and the assumption that Poisson's ratio is 0.28, leads to a Young's modulus of 1970×10^9 dynes/cm².

Attempts to detect the torsional resonant frequency were not successful.

2. Irradiations of Refractory Ceramic Fuel Materials

Clad specimens of various refractory ceramic compounds of uranium and plutonium are being irradiated over a range of burnups and temperatures to test their behavior with respect to swelling, fission-gas release, microstructural changes, and their behavior with various cladding materials.

Four instrumented, temperature-controlled capsules containing six clad specimens are being irradiated in MTR. One capsule, ANL-56-7, contains four specimens of vibratorily compacted, physically mixed powders of UC-20 w/o PuC and two specimens of vibratorily compacted PuC powder. All six specimens are clad with 0.23-mm-thick Nb-1 w/o Zr and have a fuel diameter of 3.9 mm. These specimens have achieved estimated burnups from 8.1×10^{20} to 9.4×10^{20} fiss/cc (22,000 to 25,500 MWd/MT-carbide) at cladding surface temperatures between 550 and 610°C. Target burnup for these specimens is 3.7×10^{21} fiss/cc (100,000 MWd/MT-carbide).

Capsule ANL-56-8 contains four specimens of pressed and sintered US pellets and two specimens of vibratorily-compacted physically mixed powders of UC-20 w/o PuC. The specimens are clad with 0.31-mm Nb-1 w/o Zr and the fuel diameter is 6.4 mm. The sulfide pellets are helium-bonded to the cladding. The specimens have achieved estimated burnups from 1.5×10^{20} to 2.2×10^{20} fiss/cc at maximum cladding temperatures from 535 to 650°C. Design cladding temperatures range from 550 to 650°C. Target burnup for these specimens is 5.5×10^{21} fiss/cc (187,000 MWd/MT-US or 150,000 MWd/MT-carbide).

Capsule ANL-56-9 contains a loading identical with that of capsule ANL-56-8. Specimen burnup to date ranges from 6.5×10^{19} to 9.9×10^{19} fiss/cc. Maximum cladding surface temperatures have ranged from 530 to 760°C. The capsule will be moved to a position of higher flux to achieve the desired temperature range from 800 to 850°C. Target burnup is 3.7×10^{21} fiss/cc (125,000 MWd/MT-US or 100,000 MWd/MT-carbide).

Capsule ANL-56-11 also contains a specimen loading identical with that of capsule ANL-56-8. This capsule was recently inserted in the reactor and the specimens have achieved burnups from 0.9×10^{19} to 2.2×10^{19} fiss/cc at average cladding surface temperatures from 500 to 620°C. This capsule will also be moved into a higher flux position to

reach desired temperatures of 550 to 650°C. Target burnup for the specimens is 3.7×10^{21} (125,000 MWd/MT-US or 100,000 MWd/MT-carbide).

D. Other Reactor Fuels and Materials Development

1. Polarization Studies in Liquid Metal Environment

Investigation is continuing of zirconium corrosion film-stripping techniques to permit determination of amount of zirconium metal loss during corrosion tests in highly oxygenated sodium. Results of cathodic reduction experiments in mercury-vapor glow discharge and of chemical reduction in Na-Ca liquid alloy are summarized below. Test results are noted of an electrical capacitance approach to the evaluation of film thickness as an alternative to stripping.

An unalloyed zirconium rod sample carrying a substantial amount of black surface-corrosion product from exposure to oxygen-contaminated sodium was made the cathode in a mercury-vapor glow discharge tube. The glow zone was held at 450°C, while the mercury vapor pressure was maintained in the range of 0.5 Torr by temperature control of another section near 110°C. Currents employed ranged from 1 mA/cm² for several hours up to 10 mA/cm² for nearly an hour. No appreciable reduction was evident from weight data and visual inspection. Apparatus modification to permit a higher sample temperature, and thus a higher mobility of oxygen ions in the film, may promote the desired reduction.

Chemical reduction in Na-Ca liquid alloy has been more successful. Calcium in amounts near the saturation concentration in sodium at the test temperature were employed at 500, 600, and 650°C for overnight exposures in a stainless steel autoclave. Samples of Zr-3 w/o Ni-0.5 w/o Fe and Zr-1 w/o Cu-1 w/o Fe alloy which had formed substantial amounts of black surface-corrosion product in steam were used, together with freshly ground control samples of the same material. The treatment transformed the outer surface of the corrosion product from a black, insulating (at room temperature) surface to one of a discolored, metallic appearance (after cleaning in water) that was an electrical conductor between surface probes. A slight weight loss accompanied the transformation.

The metallic layer was afterwards etched from one sample in HNO₃-HF solution, which restored a dark color and insulating quality of the surface, and caused a moderate weight loss. The metallic layer is thought to be composed of partially reduced zirconium, still rich in oxygen (based on scratch resistance): a sample has been submitted for spectrographic identification. Since the reduction reaction appears to diminish after the formation of the metallic layer, alternate exposures to Na-Ca and to etchant may be required to strip the film completely. Alternatively, the temperature might be raised to increase oxygen diffusion through the reduced layer. Control samples were somewhat discolored by exposure to Na-Ca and exhibited slight weight gains.

Useful layer composites of metal-oxide-metal may be manufactured by the above technique; however, whether the metallic layers were electrically isolated was not established.

Measurement of electrical capacitance between sample metal and the outer surface of corrosion film is being investigated as an index of film thickness. Difficulty is encountered from electrical shorts through the film. Boric acid anodization to eliminate shorts has been partially successful. However, when a silver mirror electrode was deposited chemically on a corroded and anodized sample, shorting was nevertheless evident. The investigation of the technique is continuing as an alternative to film stripping.

2. Corrosion of Iron and Nickel Alloys in Superheated Steam

Some additional metal losses from defilmed specimens have been obtained for corrosion in 750°C, 42-kg/cm² static steam (containing about 30 ppm oxygen). They are reported along with earlier data in Table XIV.

Table XIV. Corrosion in Superheated Steam

Alloy	Metal Loss - mg/cm ²					
	7 Days	14 Days	21 Days	28 Days	35 Days	50 Days
304 SS (EP)*	8.3	10.9	14.3	23.0	29.5	34.5
304 SS (GB)			3.1			
316 SS (EP)	12.2	17.8	17.7	21.9	21.5	
321 SS (EP)	12.9	17.5	25.2	24.4	31.8	
347 SS (EP)	11.9	16.7	24.8	27.9	37.5	
406 SS (EP)	0.4		0.42	0.42	0.55	
Inconel 600 (EP)	9.4	13.5	17.9	12.4	13.8	
Inconel 600 (WG)				1.8		
Inconel 625 (EP)	0.4	0.6	0.6	0.7	0.7	
Inconel X750 (EP)	2.9	2.4	2.4	2.6	3.27	
Incoloy 800 (EP)	6.7	7.2	6.8	6.8	7.2	
Incoloy 800 (GB)						2.9
Rene 41 (EP)	0.96		1.04	1.16	1.33	
Nichrome V (EP)	0.5					
302B (EP)					3.3	

*(EP) - Electropolished

(GB) - Grit Blasted

(WG) - Wet Ground 600 Grit, Light Electropolish

It is obvious that the method of preparation of sample surface, first noted at 650°C, is still an important variable in the corrosion behavior at 750°C.

Silicon (302B) reduces the corrosion of the 300 series stainless steels to a much more acceptable value. Type 406 and Inconel 625 have outstandingly good resistance to this environment, even in the poorest surface condition.

A 72-day dynamic test of Incoloy 800 modified with small additions of aluminum has been completed. Test conditions were 61-m/sec velocity, 650°C, pressure of 42 kg/cm², 30 ppm oxygen, and 3.8 ppm hydrogen. In contrast with a test run in static steam at the same conditions, there was no direct correlation between aluminum content and corrosion behavior. All five alloys performed well, particularly one containing 2.4% Aluminum.

E. Heat Engineering

1. Two-phase Flow Studies

a. Void Fraction - Pressure-drop Facility. During the past reporting period, the loop has logged more than 600 hr of continuous operation at a temperature of 750°F during wetting and calibration procedures. There is, as yet, no indication of excessive oxide accumulation, and, pending the results of 1200°F operation, it may be possible to obtain boiling data without use of the flushing and dumping procedures previously anticipated.

2. Boiling Liquid Metal Technology

a. Niobium-1% Zirconium Loop Construction. Replacement of the faulty cooling lines on the vacuum chamber was completed; however, during final cleanup of the upper chamber, additional leaks were discovered. These are now being repaired, but delivery of the loop has again been delayed. Drafts of the operation and maintenance manual were prepared and are being reviewed. A cost analysis of alternative schemes to supply liquid nitrogen and a review of expenditures for the general project are being made.

Interlocks in the plans for the main seal and cold trap, cooling-water circuits, together with the design of a cover plate for the chamber pumping port, have been included in the overall design.

b. Heater Experiments

(i) Thermal Radiation Heater Experiment. The tantalum radiation shields surrounding the thermal radiation heated section are being replaced. The loop instrumentation is being improved to obtain more reliable temperature readings in the heated section. Upon reassembly, the system shall be operated to extend the temperature level and heat flux range previously obtained.

(ii) Electron-bombardment-heating Experiment. Fabrication of equipment for the electron-bombardment-heating experiment (see Progress Report for January 1964, ANL-6840, p. 70) is complete, and the unit is being assembled. The heating system consists of a tube with heat applied to the inside surface while the liquid metal is in contact with the outside surface. Heating is accomplished by placing an electron emitter (cathode) inside the evacuated tube. The electrons liberated by heating are drawn to the inside tube surface by use of an accelerating voltage. The heat generated as a result of electron bombardment flows through the tube wall to the liquid metal.

The system being assembled is a sodium pool-boiling experiment with a 16-in. heated length. A 0.030-in.-diameter thoriated tungsten wire will be used as the cathode. Thermocouples will be placed inside the tube (anode) wall to obtain temperature measurements. Heat will be removed from the system by means of cooling coils placed above the liquid sodium level on which sodium vapor will condense. The system will be pressurized by means of an argon gas blanket.

3. General Heat Transfer

a. Heat Transfer in Double-pipe Heat Exchangers

(i) Liquid Metal Cocurrent Turbulent Flow

Experiment. Fabrication of the mercury heat transfer loop (see Progress Report for April 1964, ANL-6885, p. 54) is nearing completion. Only minor welds at various locations around the flow loop remain to be made. When completed, the loop will be pressure tested for leaks, carefully cleaned, and then filled with mercury.

Analysis. As reported in the Progress Report for June 1964, ANL-6912, pp. 72-3, an analytical solution for a liquid metal, cocurrent-flow, double-pipe heat exchanger without the narrow annular space idealization used previously has been developed. In a comparison between results yielded by this formulation and the solution based upon the assumption of a narrow annular space, it was shown that the accuracy of the approximation involving a narrow annular space decreases as the annular ratio R decreases.

For values of $K < 1.0$ the two formulations yield nearly equivalent fully developed overall Nusselt numbers for annular ratios as low as 0.1.

The analysis yields a relationship for the heat exchanger effectiveness which can be written as

$$\epsilon(z) = 1 - \phi(z) \exp \left[- \frac{1+H}{H} \text{Nu}_1^0(\infty) z \right],$$

where

$$z = \frac{2}{\text{Pe}_1} \frac{\ell}{a_1}.$$

In the above, $\text{Nu}_1^0(\infty)$ is the fully developed value of the overall Nusselt number, Pe_1 is the tube-side Peclet number, ℓ is the exchanger length, a_1 is the radius of the tube, and H is the heat capacity flow rate ratio (see Progress Report for June 1964, ANL-6912, p. 22). The quantity $\phi(z)$ is called the effectiveness coefficient and is a function of the four basic parameters H , K , K_w and R as well as the dimensionless distance value z . A comparison with the customary relation for the effectiveness shows that the two relations are equivalent when $\phi(z)$ is unity. It was shown that for a cocurrent flow exchanger $\phi(z)$ is always less than unity, and thus could be an important quantity for heat exchanger design.

The effectiveness coefficient $\phi(z)$ reaches a fully developed value, $\phi(\infty)$, which can be significantly less than unity as illustrated in Figure 25, where $\phi(\infty)$ is shown as a function of H for fixed values of R , K , and K_w .

Figure 25

Fully Developed Effectiveness Coefficient $\phi(\infty)$
vs. Heat Capacity Flow Rate Ratio H

In order to investigate the importance of the quantity $\phi(z)$, calculations were made to compare the heat exchanger length required for a specified effectiveness as predicted by the customary and exact relations. The results suggest that the customary relation based on overall Nusselt numbers corresponding to boundary conditions of uniform heat flux will give satisfactory results for effectivenesses of 0.7 and higher.

For smaller effectivenesses, the exact solution utilizing the effectiveness coefficient and actual fully developed Nusselt numbers must be used. Use of the customary relation based on Nusselt numbers corresponding to the normally assumed boundary condition of uniform wall temperature yielded unsatisfactory results for effectivenesses less than 0.9.

(iii) Cocurrent Laminar Flow. Previous analyses of the cocurrent double-pipe heat exchanger Graetz problem (i.e., with fully developed laminar flow on both sides of the exchanger) gave useful results over a limited range of operating conditions.¹⁶ The main reason for this limited range was that the problem, as formulated for evaluation by digital computer, would require an unsatisfactorily large amount of machine time to cover the entire range desired. The problem has been reformulated for solution by electronic analogue computation, and includes removal of the narrow annular space idealization used previously. During the past month, the required analogue circuitry has been checked out, and a trial case has been run with satisfactory results.

(iv) The Counter-current-flow Exchanger. Attempts to obtain a tractable mathematical formulation applicable to the countercurrent flow exchanger similar to those obtained for cocurrent flow have not, until recently, been successful. The very special case in which a uniform and constant heat transfer coefficient was assumed for one side of the exchanger resulted in a tractable formulation (see Progress Report for October 1963, ANL-6801, p. 53). A suggestion has been made for a method of attack which has resulted in a formal analytical solution for the more general case. Although further work is required, it is fairly certain that useful analyses of counter-current cases are now feasible.

b. Fog-flow Heat Transfer and Fluid Flow. As mentioned in the Progress Report for February 1964, ANL-6860, p. 71, extensions of previous mathematical descriptions of idealized flow of a highly dispersed mixture of liquid in its own vapor, or fog flow, were in progress. These extensions were described in some detail at the Joint USAEC-Euratom Two-phase Flow Meeting, Washington, D.C., April 29-May 1, 1964.

Of continuing interest is an idealization which results in the simplest mathematical representation. This representation, called the Negligible Liquid Film Model, can be related to existing experimental data exploring the occurrence of a critical heat transfer condition in a uniformly heated circular duct, but requires solutions of a nonstandard linear boundary value problem. Previous solutions to this problem were obtained by various approximation methods, the accuracy of which were somewhat uncertain when applied to the entire range of interest.

¹⁶Stein, R. P., The Graetz Problem in Co-current Flow Double-pipe Heat Exchangers, Paper presented at Seventh National Heat Transfer Conference, Cleveland, August 9-12, 1964, AIChE Preprint No. 11.

During the past month, the problem was rephrased for solution by electronic analogue computer, and results were obtained over the entire range of interest. There is good agreement with results obtained previously except, as expected, for large values of a parameter B (proportional to the heat flux) and small values of the duct length. Results were also obtained from which the effect of nonuniform liquid concentration distribution at the duct inlet on the occurrence of a critical heat transfer condition can be studied. Previous results were for uniform inlet concentration distributions only.

4. ANL-AMU Program

Other heat engineering experiments, performed as part of a joint program (not supported by the Division of Reactor Development) between the Laboratory and the Associated Midwest Universities (AMU), are described below:

a. Propagation of Void Waves in an Air-Water System. Results of the first set of test runs (see Progress Report for June 1964, ANL-6912, p. 76) were obtained from the 704 computer. From the results, it appears that a greater number of cycles are necessary in order to obtain a good statistical average of the propagating wave. Therefore, two more test runs were made, one of 500 cycles and another of 1000 cycles.

As the reliability of the electrical resistivity probe has not yet been truly established, two more experiments were performed.

One probe was soaked in a solution of sodium ethyl xanthate (a wetting agent) for 2 hr, while another was soaked in water. The difference in void fraction values obtained from the two probes was insignificant.

The cross-sectional average void fraction determined by the probe was checked against another reliable method. If air is bubbled up one side of a U-tube apparatus filled with water, the difference in fluid level height is due to the voids. The probe gave results about 14% lower than the measured values.

During the course of the test runs, it was observed that when the system was perturbed, slug bubbles were formed instead of an increased concentration of small bubbles. Granulated sugar, added to the water in amounts up to 8 lb per 100 lb water in an attempt to prevent coalescence, appeared to have little effect.

b. Power-to-void Transfer Functions. The experimental investigation of power-to-void transfer functions (see Progress Report for July 1964, ANL-6923, p. 67) has been continued with runs at 300 and 400 psia. In Figure 26, the transient void amplitude is plotted as a function of frequency for the case in which the "one-shot" Y-attenuation technique was

used to measure the void response. The theoretical curve was obtained by means of the Marchaterre-Hoglund¹⁷ correlation for two-phase flow in a digital computer program¹⁸ written following the method of Hudson, Atit, and Bankoff.¹⁹ The agreement between theory and experiment is quite good up to about 1.5 cps, at which the signal-to-noise ratio becomes less than 1 part in 20 and therefore the void amplitudes above this frequency are not too reliable.

Figure 26

Transient Void Amplitude vs. Frequency
by "One-shot" Y-attenuation Technique

Figure 27 gives the transient void amplitude as a function of frequency at various transverse positions which were obtained using a 1/32-in. window in the Y-attenuation technique. A slight two-dimensional effect is evident here in that the amplitude of the void response is higher at the centerline of the test tube than it is near the wall. However, this effect is only noticeable in the very low-frequency range from 0.01 to 0.05 cps; beyond this frequency range there is no detectable amplitude difference between the two cases.

¹⁷Marchaterre, J. F., and B. M. Hoglund, Correlation for Two-phase Flow, Nucleonics, 20 (8) (1962).

¹⁸K. Jain. Private communication.

¹⁹Hudson, J. L., K. M. Atit, and S. G. Bankoff, Response of a Boiling Channel to Power or Inlet Flow Modulation, Chem. Eng. Sci., 19, 387 (1964).

Figure 27

Transient Void Amplitude vs.
Frequency by Y-attenuation
Technique with 1/32-in. Window

The phase lag between the power and void signals is shown in Figure 28, where there is, again, good agreement between theory and experiment. There is no evident difference in the phase lag between results obtained at the centerline and those closer to the test section walls. Also, the results from the "one-shot" method agreed very well with those obtained with the 1/32-in. window.

Figure 28
Phase Lag between Power
and Void Signals

The range of linearity of the void response to power modulation has also been investigated at various frequencies at 300 psia. The results (see Figure 29) indicate that the linear range extends up to at least 19% power modulation peak-to-peak.

Figure 29
Void Response to
Power Modulation

Similar runs have also been made at 400 psia, 20 kW, and 6 gpm, with the narrow window, but the data has not yet been reduced.

F. Chemical Separations

1. Fluidization and Volatility Separation Processes

a. Recovery of Uranium from Low-enrichment Ceramic Fuels.

Laboratory work in support of the pilot-plant study of a fluid-bed fluoride volatility process is directed toward determining the optimum conditions for the fluorination of uranium-plutonium oxide mixtures in a fluidized bed of alumina granules and for efficient removal of plutonium from the fluid bed. Current experiments are directed toward determining the quantity of plutonium retained by the alumina granules after successive re-use of the bed. Low losses of plutonium to the alumina can be expected if plutonium retention does not increase with re-use of the bed. The present aim is to limit the loss of plutonium to the alumina waste to no more than one percent of the plutonium charged to the fluid-bed reactor.

In a previous report (see Progress Report for June 1964, ANL-6912, pp. 76-77), an experiment was described in which the effect of re-use of the alumina bed material on plutonium retention was measured. In this experiment, it was observed that after eight additions of a U_3O_8 - PuO_2 -fission product mixture were made, greater than 99% of the total

quantity of plutonium charged to the reactor was volatilized. This experiment did not include molybdenum oxide (MoO_3) among the selected fission product oxides since its use had resulted in operational problems associated with plugging of lines in the product collection manifold.

In recent work it was observed that the presence of MoO_3 in the U_3O_8 - PuO_2 -fission product feed material affected the course of the volatilization of plutonium during several re-uses of alumina bed material. Five additions of U_3O_8 - PuO_2 -fission product mixture containing 1.2 g plutonium, 254 g uranium, and 2.58 g of selected fission product oxides (La_2O_3 , CeO_2 , Pr_6O_{11} , Nd_2O_3 , Sm_2O_3 , Eu_2O_3 , Gd_2O_3 , Y_2O_3 , BaO , ZrO_2 , and MoO_3 ; the concentration of MoO_3 was 22% of the fission product oxide mixture) were made to a single alumina bed. For each addition, 300 g of the oxide mixture was blended with 150 g of alumina and injected into the alumina bed at 450°C where the material was reacted with 20 v/o fluorine in nitrogen. The solids feed rate during the feeding-fluorination period was approximately 2.3 g U_3O_8 /min. Following the feeding-fluorination period, the fluorination was continued with 100% fluorine with continuous recycle at three temperature levels: 450°C for 5 hr, 500°C for 5 hr, and 550°C for 10 hr. At the end of each complete cycle, the alumina bed was sampled for plutonium analysis and then divided into two parts, 400 g to be used for the fluid-bed material, and 150 g to be mixed with the next batch of oxide feed.

After the first addition-fluorination cycle, the alumina bed contained 0.003 w/o plutonium. The plutonium concentration of the alumina rose to 0.030 w/o after the second addition-fluorination cycle, and then gradually decreased to 0.014 w/o after the fifth cycle. The concentration of 0.014 w/o plutonium on the alumina after the fifth cycle corresponds to volatilization of 98% of the plutonium charged to the reactor. If the plutonium concentration remained at this level for two additional cycles, then the alumina should contain an amount of plutonium equivalent to less than 1% of the plutonium charged to the reactor.

Comparison of these results with results of the test in which MoO_3 was not present shows that the presence of MoO_3 affects the course of plutonium volatilization. In the absence of MoO_3 , a plutonium concentration on the alumina bed equivalent to 1% of the total plutonium charged to the reactor is achieved after two addition-fluorination cycles, whereas, in the presence of MoO_3 , it is likely that the quantity of plutonium retained on the alumina can be reduced to 1% of that charged to the reactor after seven addition-fluorination cycles. Seven re-use cycles are not only economically attractive, but also appear feasible on a process basis.

In support of the work described above, preliminary experiments have been carried out which show that at 450°C the reaction of MoO_3 and fluorine, diluted with nitrogen or oxygen, proceeds with the

formation of high-boiling material, which is probably MoOF_4 , MoF_5 , or a mixture of the two. Only a small fraction (3 to 5%) of the molybdenum is converted to molybdenum hexafluoride (MoF_6). No reaction was observed to occur between MoF_6 and oxygen at 450°C .

b. Engineering-scale Alpha Facility. An engineering-scale alpha facility is being installed to permit demonstration of the major steps of the fluid-bed fluoride volatility process for the recovery of uranium and plutonium from ceramic oxide fuels (see Progress Report for December 1963, ANL-6810, p. 38). Shakedown testing of the fluorinator system is underway with the major objectives being a) to investigate the performance of the various equipment components, b) to demonstrate satisfactory removal of uranium from the UO_2 charge and satisfactory collection of the UF_6 product in refrigerated condensers, and c) to determine accurate uranium material balances.

The first shakedown test involved the two-zone oxidation-fluorination of a 3-in.-deep (2.2 kg) bed of UO_2 pellets in a 3-in.-dia nickel reactor. The pellets were immersed in a 24-in.-deep bed of alumina. The uranium dioxide pellets were completely converted to UF_6 in 9 hr. The bed temperature was maintained at 450°C for the initial 6.5 hr, and then raised to 500°C for the remainder of the test. The residual uranium content of the alumina bed at the end of the test was 0.03 w/o, equivalent to 0.07% of the uranium charged to the reactor. Subsequent refluorination of a portion of the alumina under static-bed conditions (1 hr at 500°C followed by 1 hr at 550°C) reduced the uranium concentration to less than 0.01 w/o.

Greater than 98% of the uranium charge was collected in the UF_6 product collection system (a refrigerated condenser backed by a sodium fluoride sorbent trap). An overall uranium material balance of greater than 99.7% was achieved. Equipment operation was considered excellent; all major components and control systems performed within design capabilities.

Installation of the 2-in.-dia converter system for the conversion to mixed oxides of the mixed UF_6 - PuF_6 product from the fluorinator system is proceeding.

c. Cleanup of Cell Exhaust Air Contaminated with Plutonium Hexafluoride. A program investigating the cleanup of exhaust air from alpha facilities used in fluoride volatility work is directed toward the design of efficient gas-cleanup systems for the removal of UF_6 and PuF_6 from glovebox atmospheres. It has been previously found (see Progress Report for April 1963, ANL-6717, pp. 53-54) that the release of pure PuF_6 or UF_6 into moist air results in reaction of the hexafluoride with the water to produce a solid reaction product (PuO_2F_2 or UO_2F_2) which is effectively removed from the air by high-efficiency (AEC) filters.

In experiments in which UF_6 was diluted with dry nitrogen prior to release into moist nitrogen (reported in ANL-6717), the hydrolysis reaction was not as rapid as when pure UF_6 was released into moist nitrogen. Additional experiments with dilute UF_6 gas streams indicate that the reaction between diluted UF_6 and moist nitrogen (p_{UF_6} between 0.000314 and 0.0308 mm Hg; $p_{\text{H}_2\text{O}}$ between 0.05 and 5.1 mm Hg) occurs primarily on solid surfaces rather than in the gas phase, as was observed when pure UF_6 was released into moist nitrogen. The rate of hydrolysis was found to increase with an increase in the partial pressure of either water or UF_6 . The rates (r) ranged from 0.3×10^{-4} to $0.1 \text{ mg U}/(\text{hr})(\text{sq cm})$ and can be correlated by the following semi-empirical relation based on an assumption that the rate-controlling mechanism is the adsorption of the hydrogen fluoride product:

$$r = \frac{191.5p_{\text{UF}_6}p_{\text{H}_2\text{O}}}{1 + 597p_{\text{UF}_6} + 95.5p_{\text{HF}}}$$

where p represents the partial pressure of the component in mm Hg. A computer program was employed to estimate the constants in the rate equation based on the results from laboratory tests; this equation correlates the data to a standard deviation of 26%.

2. General Chemistry and Chemical Engineering

a. Studies of Mobile Blanket Fuels. An evaluation of the advantages and disadvantages of mobile blankets and of various potential mobile blanket materials for fast breeder reactors, considered as alternatives to solid blankets, has begun.

In solid blanket materials, the neutron flux and the plutonium concentration are highest near the core. As a result, the high heat flux in the blanket region near the core may necessitate removal of blanket elements having a low average plutonium concentration, resulting in high rates and costs for blanket processing. Also, the plutonium buildup in the outer regions of the blanket is so slow that processing of this material is uneconomical in some cases. Although the outer regions of a solid blanket may be moved toward the center of the core to increase their plutonium concentration, this action may require reactor shutdown. An effective mobile blanket would eliminate blanket refabrication costs and variation of the plutonium concentration in the blanket.

Experimental work has been initiated to develop a paste blanket fuel consisting of UN or UC dispersed in sodium, lead, bismuth, or their alloys. Pastes appeared to be more attractive than molten salts or liquid metal alloys containing dissolved fertile material.

Because sodium has a good thermal conductivity and a low rate of corrosion of proposed reactor construction materials, sodium is an attractive dispersing vehicle for pastes to be used as fast reactor blankets. The wetting properties of ceramic blanket materials by liquid sodium are of importance in choosing a mobile blanket system. Preliminary wetting experiments have been done by the sessile drop method,²⁰ in which the contact angle between a drop of sodium and a plaque of the solid under investigation is determined by direct measurement on a photograph and by calculation from drop dimensions. By both measuring techniques, the contact angle for oxygen-saturated sodium on UO_2 was approximately 131° at about 210°C . The value obtained for oxygen-saturated sodium by Bradhurst and Buchanan²¹ was $\sim 150^\circ$; Livey and Murray²² obtained a value of 145° at 200°C . Thus, the preliminary results are in fair agreement with these literature values.

b. Preparation of Plutonium Carbide-Uranium Carbide Solid Solution. Three runs have been completed in which the objective was to prepare a solid solution of the monocarbides of plutonium and uranium, $[\text{U}_x\text{-Pu}_{(1-x)}]\text{C}$. Approximately 9 g of uranium metal and 2 g of plutonium metal were dissolved in molten zinc contained in a graphite crucible, at approximately 630°C , while in contact with a $\text{LiCl-MgCl}_2\text{-ZnCl}_2$ molten salt solution. The ZnCl_2 oxidized the uranium and plutonium metal to the trichlorides, which then transferred to the salt phase. At the completion of this reaction, the solutions were allowed to cool. The salt phase containing the heavy metal chlorides was mechanically separated from the metal phase and placed in a tantalum crucible along with magnesium sesquicarbide. The carbide-precipitation reaction was carried out at approximately 625°C for one hour. Then the salt was separated from the precipitated carbide by distilling in vacuum at 850°C . The carbide was recovered as a solid residue from the tantalum crucible.

In each of the three runs, the oxygen content of the product was about one percent, which is higher than is desirable. The products from the first two runs exhibited a face-centered cubic crystal structure and lattice parameters of either $4.959 \text{ \AA}$ or $4.960 \text{ \AA}$. The lattice parameter for UC is $4.960 \text{ \AA}$ and is the value that would be expected for solid solutions of (Pu-U)C containing up to 15 a/o plutonium. The precipitated products of the first two experiments contained 15 and 27 percent of the plutonium charged, whereas essentially all of the uranium had precipitated. Analysis of the salt phase from the third run indicated that greater than 99 percent of the uranium and plutonium originally present had precipitated in the salt phase; the fused salt has been removed from the carbide by distillation, and analysis of the carbide product is underway.

²⁰W. D. Kingery and M. Humenik, Jr., J. Phys. Chem. 57, 359 (1953).

²¹D. H. Bradhurst and A. S. Buchanan, Australian J. Chem., 14, 397 (1961).

²²D. J. Livey and P. Murray, Plansee Proc., 1955, 375.

3. Chemical-Metallurgical Process Studies

a. Chemistry of Liquid Metals. A preliminary value for the activity coefficient of plutonium in liquid magnesium has been computed from data on the partition of plutonium between a molten salt (30 m/o NaCl-20 m/o KCl-50 m/o MgCl_2) and zinc-magnesium solutions. The value was 9.2 ± 0.9 for the temperature range 600-800°C in a solution containing 0.08 a/o Pu. This datum will be useful in the design of fuel-reprocessing methods in which metallic solutions containing magnesium are used.

b. Thermodynamics of Rare Earth-Cadmium Systems. The thermodynamics of selected rare earth-cadmium systems is being studied by the effusion method. Knowledge of the eutectic temperatures for these systems and of the decomposition pressures in the MCd-M (M = rare earth) regions is needed to measure the standard free energies of formation of intermetallic compounds of these systems. Provisional values of the eutectic temperatures in the MCd-M regions of the La-Cd, Ce-Cd, Pr-Cd, Nd-Cd, and Sm-Cd systems (see Progress Report, June 1964, ANL-6912, p. 82) and of the Gd-Cd system (719°C) were previously obtained by differential thermal analysis.

A value of 458°C has been obtained by differential thermal analysis for the eutectic temperature in the Yb-Cd system. This value is within the temperature range predicted by analogy with the six rare earth-cadmium systems previously studied.

Preliminary data have been obtained on the decomposition pressures in the LaCd-La, PrCd-Pr, and NdCd-Nd regions. Very low decomposition pressures (10^{-4} to 10^{-5} Torr) were found in these systems.

G. Plutonium Recycle Program

1. EBWR Plutonium Recycle Experiment

Physics parameters for the initial EBWR plutonium loading and the plutonium criticals have been determined by the calculational model described previously (see Progress Report for July 1964, ANL-6923, pp. 75-8). Cross sections for various nuclides were determined separately for each of the four core regions. In previous calculations cross sections for all nuclides had been obtained for the spectrum characteristic of the central plutonium region. A comparison of various parameters calculated by the two methods is given in Table XV.

A further test of the calculational model is now being made using the experimental data from Hanford's subcritical experiments with 1.82 w/o Pu-Al rods.

Table XV. Comparison of Physics Parameters for the EBWR Plutonium Loading as Calculated by Two Methods at Zero Burnup

	Present Model	Previous Model
k_{eff} (at 42 MW)	0.995	1.000
k_{eff} (cold, clean)	1.144	1.152
k_{eff} (Plutonium Region)	1.085	1.085
Void Coefficient of Reactivity, $\delta k/\delta v$, (cold, clean)	-0.124	-0.10
Void Coefficient of Reactivity, $\delta k/\delta v$, (Plutonium Region, unpoisoned)	-0.194	-0.18
Boric Acid Coefficient of Reactivity, $\delta k/g/\text{gallon}$, (cold, clean)	-0.012	-0.011

2. Plutonium Recycle Fuel

Fabrication of the fuel loading for the Plutonium Recycle Experiment (see Progress Report for May 1964, ANL-6914, pp. 91-3) is a joint Argonne-Hanford program. The fuel loading is a three-zone loading with plutonium in the central zone, partly enriched uranium in the intermediate shim zone, and natural uranium in the outer blanket zone. All fuel elements are designed to contain 36 fuel rods in a square lattice. Each fuel rod contains oxide fuel in 0.945-cm-ID x 0.064-cm-wall Zircaloy-2 tubing. The loading and composition of fuel for each zone is as follows:

(a) The central zone of 36 elements is fueled with 1.5 w/o plutonium oxide and 98.5 w/o depleted uranium oxide. The loaded fuel rods for these elements are being supplied by General Electric, Hanford, for loading into elements at ANL.

(b) The intermediate zone of 60 elements is fueled with UO_2 enriched to 6 w/o U^{235} . This fuel also contains a mixed burnable poison, 0.158 w/o Eu_2O_3 and 0.0288 w/o Sm_2O_3 , uniformly distributed throughout the fuel. This fuel is being fabricated on Contract 31-109-38-1622 by United Nuclear Corporation.

(c) The blanket zone of 58 elements is fueled with natural UO_2 . This fuel is also being fabricated on Contract 31-109-38-1622 by United Nuclear Corporation.

The PuO_2 - UO_2 fuel for the central zone was prepared at Hanford by the following process steps:

- (1) Convert plutonium to PuO_2 .
- (2) Blend PuO_2 with UO_2 and compact by means of a high-energy rate-forming, "Nupac," process using Dynapak equipment.

(3) Crush and screen Nupac fuel and load into fuel rods by vibratory compaction, "Vipac" process, using vertical excitation of the tube.

(4) Inspect, clean, and test loaded rods by conventional procedures.

The required total of 1512 central-type Plutonium Recycle fuel rods acceptable for reactor service have been loaded. A total of 810 completed fuel rods have been delivered to ANL for loading into fuel elements.

The stainless steel upper support grids for plutonium fuel rods and stainless steel lower support grids delivered to ANL by United Nuclear Corp. (see Progress Report for July 1964, ANL-6923, p. 74) have been rejected as not conforming to specifications and will be replaced. Center spacer grid and fuel can components have not been received for evaluation.

All the 6% enriched UO_2 pellets containing the rare earth burnable poison have been fabricated, tested, and inspected. These pellets have been assembled into weighed stack heights ready for loading into fuel jacket tubing.

3. Plutonium Recycle Control Rods

Eight new control rods and one spare central oscillator rod are being fabricated for the Plutonium Recycle Experiment. The eight control rods have boron-stainless steel absorber sections with 2 w/o boron enriched to 90 w/o B^{10} contained in Type 304 stainless steel. The absorber section of the oscillator rod consists of hafnium.

The boron-stainless steel was fabricated by Carpenter Steel Co. in accordance with ANL specifications. A total of 34 pieces of strip stock were finished to 0.33 x 28 x 150 cm (0.130 x 11 x 60 in.).

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Core Physics

Two principal physics studies were continued: a) burnable poisons for long-term reactivity control, and b) a combination of burnable-poison sleeves and a revised fuel loading to permit the use of a less severely graded fuel loading without too large a penalty in the peak "thermal" flux in the internal thermal column.

a. Burnable Poisons in the Reactor Core. In the case of a burnable poison which is uniformly distributed in the fuel, several individual chemical elements have been studied, namely, boron (B^{10}), samarium, and europium. Boron would be quite interesting if some way could be found to maintain fuel-plate integrity when 70% or more of the boron (B^{10}) has been lost to neutron capture. Samarium burns out too rapidly to be of interest by itself. On the other hand, europium does not burn out rapidly enough in spite of its high microscopic capture cross section, because neutron capture results in production of other europium isotopes with high cross sections. Europium, therefore, acts more as a permanent rather than a burnable poison. The program of calculations will be continued to examine the utility of using different burnable poisons in different parts of the core, and to study a rapidly narrowing field of other elements of conceivable value.

The case of lumped samarium as a burnable poison shows greater promise. Lumped B^{10} is of little interest because even when uniformly distributed, too much boron is left unburned. For the present concept of a core with a graded fuel loading, the use of several annular zones of a cermet of steel and samarium oxide over the central region of the core is sufficiently promising that analytical burnup studies will be made.

b. Burnable-poison Sleeves. Calculations have been made for a sleeve of a neutron absorber at the inner surface of the core and with a uniform instead of a graded fuel loading. If the sleeve composition and thickness are chosen so as to retain a higher-than-average power in the core region immediately adjacent to the internal thermal column too many thermal neutrons penetrate to the core, and the problem of a steep power gradient remains. Simultaneously, the sleeve reduces reactivity; this behavior is unacceptable unless the sleeve itself acts like a burnable poison. A sleeve containing boron is inadmissible at least because it would burn up too rapidly. Other candidates are hafnium and rhodium, which have desirable thermal and near-thermal cross-section characteristics.

This study is continuing to find a sleeve which will offer a desirable burnup characteristic and will permit the use of a less severely

graded fuel loading without an unacceptable reduction in the fluxes over the core cycle available for experiments.

2. Critical Experiment

The initial fuel loading of the AARR Critical Experiment has been delayed because of lack of fuel. In addition, the welding fixtures required reworking before satisfactory welding of stainless steel envelopes over the fuel was obtained. The electron-beam welder required repairs, and the backlog of work to be done with this welder continued to grow.

Delivery of bare uranium foil slipped farther behind the expected rate; however, there is no immediate use for the foil since the stainless steel envelopes have not yet been welded over the supply on hand. Continued slow delivery of bare uranium may result in delays in loading the reactor, since successive core loadings require substantially larger amounts of fuel.

Final checkout of the facility commenced following receipt of the twelve hafnium control blades. Arrangements are being made for the AEC pre-startup survey to start after final checkout.

The revised Safety Analysis Report (ANL-6929 Draft) has been issued. The Operating Manual has been approved in draft form and copies are being issued.

Current efforts are directed toward finding a way of reducing air entrapment as moderator is raised in the fine-structured fuel assemblies. Work with dummy assemblies has indicated sufficient retention of air to affect moderator content significantly. Tests with wetting agents in the moderator gave no indication of improvement by this approach.

3. Selection of Architect-Engineer

Proposals have been received from thirteen of the sixteen architect-engineer firms solicited in connection with performance of design work on AARR. The remaining three firms declined to propose because of existing work commitments. The proposals are under evaluation by the Contractor Selection Board.

B. Magnetohydrodynamics (MHD)

1. MHD Power Generation - Jet Pump Studies

The effects of steam-nozzle pressure, upstream-liquid temperature, and downstream static pressure on the steam-nozzle efficiency, the combiner efficiency, and the overall efficiency of the jet pump have been determined by analyzing the experimental data obtained from jet-pump operation.

In addition to the results obtained from semi-closed-cycle operation of the jet pump in the pumped loop, measurements of steam flow rate and thrust for the converging-diverging steam nozzle were required to complete the analysis. The efficiency of the converging-diverging steam nozzle (divergence angle of 25°) was found to vary between 82% and 96%, depending upon the inlet steam pressure. The maximum overall efficiency of the jet pump (defined as the ratio of the actual kinetic energy of the liquid to the energy supplied by the steam expanding ideally through the nozzle) was determined to be 4.91%. Inefficient operation of the combiner section, where turbulent mixing of the liquid and vapor occurs, appears to be responsible for the major loss of kinetic energy within the jet pump.

The jet pump was first operated with a converging-diverging nozzle having a 25° angle of divergence, then with a nozzle having a 7.5° angle of divergence. The effect of the nozzle divergence angle on the performance of the jet pump is indicated in Table XVI.

Table XVI. Performance of Jet Pump

	Comparison A		Comparison B		Comparison C	
Nozzle divergence angle, degrees	25	7.5	25	7.5	25	7.5
Steam inlet pressure, psig	80	80	103.5	98	60	60
Loop upstream static pressure, psig	6	13.5	9	20	-4.2	7
Loop downstream static pressure, psig	13	19	25	25	11.2	11.5
Loop flow rate, gpm	11.4	12.1	12	13	9.6	11.2
Overall jet pump efficiency, %	3.52	4.23	2.91	4.47	3.08	3.09

The results indicate that the overall efficiency of the jet pump is improved by utilizing the steam nozzle having the smaller angle of divergence. Note the 50% increase in efficiency for the conditions of Comparison B.

The overall efficiency of the jet pump appears to be strongly affected by the temperature of the inlet liquid. During one run the system flow rate increased from 8.75 gpm to 11 gpm, an increase of 25%, as the inlet liquid temperature was decreased from 148°F to 58°F . The overall efficiency of the jet pump was 1.3% (148°F inlet, 8.75 gpm) and 2.62% (58°F inlet, 11 gpm). This increase in efficiency was not unexpected because the condensation process becomes more effective as the inlet temperature decreases. If subcooling the inlet liquid would produce a 25% increase in system flow over the entire operating range, the present 13-gpm flow rate at the

optimum operating condition (without subcooling) would rise to 16.25 gpm. Because the overall efficiency of the jet pump is a function of the square of the system flow rate, it is conceivable that the efficiency of the current configuration of the jet pump could be increased to 7.22% by subcooling the inlet liquid.

Liquid injection in the steam nozzle is currently being investigated to determine its effect on jet-pump overall efficiency and loop performance.

C. Regenerative EMF Cells

1. Bimetallic Cells

a. Sodium-Bismuth Cells. The ternary eutectic NaF-NaCl-NaI (m.p. about 530°C) is being studied for possible use as the electrolyte for the sodium-bismuth cell. In most of the previous studies of the sodium-bismuth cell, the NaCl-NaI eutectic was used. Fluoride was added to the binary eutectic since the fluoride may be effective in preventing metal dispersions in the electrolyte. Preliminary work has tentatively established the composition of the ternary eutectic as 54.6 m/o NaI-28.7 m/o NaCl-16.7 m/o NaF.

Provisional solubilities of sodium metal in the ternary eutectic range from 0.3 m/o at 546°C to 0.9 m/o at 697°C. These results may be low because of sodium losses by volatilization. Additional solubility experiments will be carried out in which an attempt will be made to minimize sodium losses.

2. Lithium Hydride Cell

Previous determination (see Progress Report for May 1964, ANL-6904, p. 101) of the standard emf of the lithium hydride cell:

has made possible the calculation of the activity of LiH in similar cells. Emf measurements were made for two LiH cells in which the compositions were 62.29 m/o LiCl-26.70 m/o LiF-11.01 m/o LiH and 66.06 m/o LiCl-28.30 m/o LiF-5.64 m/o LiH. From these data, the activity of LiH at 540°C (the temperature at which the standard emf had been determined) was calculated for each electrolyte. For electrolyte compositions of 11.01 m/o LiH and 5.64 m/o LiH, the activities were calculated to be 0.41 and 0.23, respectively, higher than expected. Additional experiments will be made to determine the role of the composition of the electrolyte with regard to the LiH activity.

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal-Water Reactions

a. Laser-beam Heating Applications. Additional experiments were carried out by the laser beam heating method to determine the effect of pressure on the aluminum-water reaction. In previous experiments (see Progress Report for July 1964, ANL-6923, p. 86), 2 x 2 x 0.025-mm foils were exposed to laser beams while submerged in room-temperature water at a total pressure of about 25 mm. The extent of the aluminum-water reaction ranged between 9 and 15%. In the present studies, two types of experiments were performed with aluminum foils of the same size. In the first type, aluminum foils were exposed to laser energy while submerged in room-temperature water with an overpressure of 1 atm of argon. In the second type, the foils were submerged in water heated to 100°C with an overpressure of 1 atm of steam.

In two experiments of the first type, the values for the extent of the aluminum-water reaction were 11 and 17%, not significantly different from the results obtained previously. In four experiments of the second type, the average value for the extent of reaction was 42%. Because extensive but variable break-up of the foils into metal spheres occurred during the heated-water experiments, the values for the extent of reaction varied widely among the tests. However, these preliminary results do indicate that the extent of the aluminum-water reaction increases with increasing vapor pressure of the water.

b. Studies in TREAT of Uranium Dioxide Fuel. Two experiments were performed in TREAT with UO_2 fuel specimens to obtain kinetic data under conditions simulating a power excursion. One experiment (CEN 187) was carried out with a Zircaloy-2-clad, UO_2 -core fuel specimen (see Progress Report for June 1964, ANL-6912, pp. 90-94, for previous results). The other experiment (CEN 188) was carried out with an unclad UO_2 pellet. In both experiments, the test specimens were exposed to TREAT transients while submerged in water at room temperature.

The data from the two experiments are summarized in Table XVII. The energy input in CEN 187 corresponded to a maximum temperature (adiabatic) of the UO_2 of 2700°C (melting temperature of UO_2). The specimen retained its integrity, and 10.2% of the cladding reacted with water. In experiment CEN 188, the energy input corresponded to an adiabatic temperature of 3300°C (vaporization temperature). Analysis indicated that the UO_2 was oxidized to a composition in the range between $\text{UO}_{2.12}$ and $\text{UO}_{2.17}$. Further experiments are being performed to verify the finding that a significant reaction of UO_2 with water occurs at high temperatures in the absence of cladding metal.

Table XVII. TREAT Metal-Water Experiments with UO₂ Fuel

	<u>CEN 187</u>	<u>CEN 188</u>
<u>Reactor Characteristics</u>		
Integrated Power, MW-sec	239	327
Peak Power, MW	1080	1130
Period, msec	74	74
Computed Energy Input, cal/g	216	409
<u>Fuel Pin</u>		
Core	UO ₂	UO ₂
Cladding	Zr-2	None
Enrichment, %	11.2	11.2
<u>Results</u>		
% of Reaction Based on H ₂ Evolved	10.2 (Zr + H ₂ O)	21.4 (UO ₂ + H ₂ O) ^a
Peak Pressure Rise, psi	9	75
Final Appearance of Fuel Pin	Intact	Many fine particles
d _{sv} , ^b mils, Mean Particle Size	375	13.5

^a Based on complete conversion of UO₂ to UO₃.

$$^b d_{sv} = \frac{1}{\sum_{i=1}^N w_i/d_i}, \text{ where } d_i = \text{average diameter of the } i^{\text{th}} \text{ size group,}$$

w_i = fraction by weight of particles in the i^{th} group, and N is the number of size groups.

B. Fast Reactor Safety Studies

1. In-pile Oxidation Experiments

In-pile oxidation studies at TREAT are being made to determine the particle-size distribution that occurs when reactor fuels are heated in flowing air in a reactor environment.

In the ninth of the series of experiments, the sample, suspended in the center of a beryllia crucible, was contained in a stainless steel capsule placed in the center of the TREAT reactor core. Within the capsule was also a series of successively finer stainless steel screens to remove the oxide particulates from the effluent air stream. Plant air, dried by silica gel adsorber, was passed over the sample and then down through the series of screens. Hole sizes of the screens were 74, 43, 25, and 5 μ , respectively. The final screen, a millipore MICROTUBE, was capable of retaining 1- μ particles. A liquid nitrogen-cooled charcoal trap and an AEC absolute filter installed in the exit gas stream provided additional protection.

After each test, the capsules were disassembled. The particles on each screen were chemically analyzed by either fluorimetric or colorimetric methods. In each test, the total recovery of uranium was at least 99%.

The samples used in these tests were either pure uranium (5 w/o enriched) or a uranium-5 w/o fission alloy. The 5 w/o fission portion contained 2.5 w/o Mo, 1.5 w/o Ru, 0.50 w/o Pd, 0.30 w/o Rh, and 0.20 w/o Zr. The pure uranium pins were 0.55 in. long by 0.10 in. in diameter, and weighed approximately 2 g. The uranium-5 w/o fission alloy pins were 0.27 in. long by 0.14 in. in diameter, and weighed approximately 1.3 g.

The summary of experimental results shown in Table XVIII indicates that with energy inputs high enough to vaporize a significant portion (15 to 30%), there is no substantial formation ($\leq 0.25\%$) of particles smaller than $5\ \mu$. To assure that no particles smaller than $1\ \mu$ escaped from the capsule exit stream, several analyses were made on the charcoal trap used as the backup. No uranium was found in any of the tests on the charcoal trap. In general, with the uranium-5 w/o fission pins, as the energy input was increased, there was also an increase in the production of particles smaller than $25\ \mu$.

Table XVIII. Summary of In-pile Oxidation Experiments

Sample		Fission Energy Input of Sample (cal/g)	Sample Left in Crucible Area (w/o)	Final Sample Distribution on Each Screen (w/o)				
No.	Type			74μ	43μ	25μ	5μ	1μ
3	U-Fission ^a	136	96.3	2.38	0.92	0.38	0.02	≤ 0.01
4	Pure U ^b	216	95.0	4.78	0.39	0.20	0.04	≤ 0.01
5	Pure U ^b	375 (30%) ^c	98.9	0.74	0.14	0.16	0.05	≤ 0.01
6	Pure U ^b	375 (30%) ^c	98.9	0.72	0.11	0.17	0.075	≤ 0.01
7	Pure U ^b	350 (28%) ^c	91.4	6.88	0.86	0.61	0.20	0.003
8	U-Fission ^a	257 (15%) ^c	96.2	2.10	0.70	0.71	0.21	0.11
9	U-Fission ^a	312 (24%) ^c	96.4	1.98	0.53	0.31	0.55	0.25

^a20% enriched uranium-5 w/o fission.

^b5% enriched pure uranium.

^cPercent volatilized.

NOTE: Air flow for each test was 10 cfm or a superficial velocity of 8 ft/sec across the sample.

C. TREAT

1. Large TREAT Loop

The shipping cask for the test section of the Large TREAT Loop has been received and checked for dimensions and specifications. The bottom plate was distorted and dished out of flat by approximately 0.084 in. In order to insure that the bottom plate of the cask would be flat and perpendicular to the cask center line, the cask was placed on the bed of a large horizontal boring and milling machine in Bldg. 363, the cask center line optically aligned with the spindle of the machine, and several facing cuts taken across the base.

Modifications have been made to the bottom drawer of the cask and the drawer cavity to improve drawer operation and insure positive sealing of the aluminum gasket. A radiation survey of the cask will be performed to check the uniformity and continuity of the lead shielding.

Additional bellows for the Large TREAT Loop have been received and have passed a mass-spectrometer test, confirming the leak-tightness data supplied by the vendor. Several bellows are being reworked after a radiographic examination indicated faults in the welds.

Additional electrical heaters for the loop were received from the supplier. Heating elements for the tanks were not included in the shipment; thus, completion of the loop assembly is delayed pending receipt of these heaters.

Welding of pipe subassemblies continued in preparation for loop assembly.

VI. PUBLICATIONS

Papers

CRYSTALLINE AMMONIUM URANIUM PEROXY-OXALATE

Yehuda Baskin and N. S. K. Prasad

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Phys. Letters 11, 4 (1964)

STATISTICAL THEORY OF NUCLEAR COLLISION CROSS SECTIONS

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SUPERHEAT FUEL CLADDING DEVELOPMENT

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STRUCTURAL MATERIAL DEFECTS

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CONTAMINATED SODIUM

H. A. Levin and Sherman Greenberg
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POLARIZATION STUDIES IN LIQUID METAL ENVIRONMENT

C. A. Youngdahl and Sherman Greenberg
p. 199C

RATE OF SOLUTION OF ROTATING TANTALUM DISKS IN LIQUID
TIN

T. F. Kassner
p. 199C

CORROSION OF STEEL AND TANTALUM BY MOLTEN CADMIUM-
MAGNESIUM-ZINC SOLUTIONS

M. L. Kyle, P. A. Nelson, and Leslie Burris, Jr.
p. 199C

DIFFUSIVITY OF URANIUM IN VARIOUS LIQUID METAL SOLUTIONS

H. E. Hootman, J. C. Hesson, and Leslie Burris, Jr.
p. 199C

CORROSION OF REFRACTORY METALS BY ZINC-MAGNESIUM-
URANIUM AND HALIDE-SALT SYSTEMS

P. A. Nelson, M. L. Kyle, G. A. Bennett, and Leslie Burris, Jr.
p. 200C

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THERMODYNAMICS OF PLUTONIUM, THORIUM AND URANIUM
METALLIC SYSTEMS

Irving Johnson
pp. 171-192

CONSTITUTION AND PROPERTIES OF PLUTONIUM MONOCARBIDE

O. L. Kruger
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PROPERTIES OF URANIUM MONOPHOSPHIDE, URANIUM MONOSULFIDE AND UP-US SOLID SOLUTIONS

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IRRADIATION EFFECTS IN URANIUM SULFIDE

L. A. Neimark and Richard Carlander

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EXTRACTION OF FISSION PRODUCTS FROM MOLTEN URANIUM-PLUTONIUM ALLOYS BY HALIDE SALTS

G. A. Bennett, L. Burris, Jr., and R. C. Vogel

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THE HEAT OF FORMATION OF SILICON CARBIDE BY FLUORINE BOMB CALORIMETRY

Elliott Greenberg, Carol A. Natke, and Ward N. Hubbard

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ABSORPTION SPECTRA OF ALKALI METAL TELLURIDES IN MOLTEN ALKALI HALIDES

D. M. Gruen, R. L. McBeth, M. S. Foster, and C. E. Crouthamel

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PILOT PLANT DEVELOPMENT OF A FLUID-BED FLUORIDE VOLATILITY PROCESS FOR HIGH ENRICHED URANIUM-ALLOY FUELS

John T. Holmes and John J. Barghusen

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REACTION OF FLOWING STEAM WITH REFRACTORY METALS.
I. MOLYBDENUM 1100-1700°C

Martin Kilpatrick and Stanley K. Lott

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BENCH-SCALE DEVELOPMENT OF A FLUID-BED VOLATILITY PROCESS FOR HIGH-ENRICHED URANIUM-ALLOY FUELS

D. Ramaswami, N. M. Levitz, and A. A. Jonke

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SMALL-SCALE DEMONSTRATION OF THE MELT REFINING OF HIGHLY IRRADIATED EBR-II URANIUM-FISSIUM FUEL

V. G. Trice and R. K. Steunenberg

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ANL-Reports

- ANL-6435 THE U-Pu-C TERNARY PHASE DIAGRAM BELOW
50 ATOMIC PERCENT CARBON
S. Rosen, M. V. Nevitt, and A. W. Mitchell
- ANL-6703 EBWR TEST REPORTS (100-MWT OPERATION)
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- ANL-6807 HEAT AND MASS TRANSFER TO A GAS-FLUIDIZED
BED OF SOLID PARTICLES
Edward N. Ziegler
- ANL-6831 NUCLEAR CRITICALITY CONTROL IN THE EBR-II FUEL
CYCLE FACILITY
J. C. Hesson and L. Burris, Jr.
- ANL-6835 FREE CONVECTION OF A LOW PRANDTL NUMBER
FLUID IN CONTACT WITH A UNIFORMLY HEATED
VERTICAL PLATE
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- ANL-6852 THE PERIOD EFFECT IN REACTOR DYNAMICS.
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- ANL-6862 TWO-PHASE HEAT TRANSFER WITH GAS INJECTION
THROUGH A POROUS BOUNDARY SURFACE
A. A. Kudirka
- ANL-6876 THE FEASIBILITY OF CENTRIFUGAL CASTING FOR
MANUFACTURING MASURCA ELEMENTS
R. L. Lesser and J. E. Ayer
- ANL-6881 AN INTRODUCTION TO MULTICHANNEL ANALYZERS
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Ed. by Leonard E. Link

Published by U. S. Atomic Energy Commission, Division of
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BOILING "BURNOUT" FOR REACTOR DESIGN

R. P. Stein and P. A. Lottes

pp. 131-176

BEHAVIOR OF CLADDING MATERIALS IN WATER AND STEAM
ENVIRONMENTS

Sherman Greenberg

pp. 213-330

PYROPROCESSING OF REACTOR FUELS

R. Dean Pierce and Leslie Burris, Jr.

pp. 411-476

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Chapter 8, "Control and Instrumentation"

W. C. Lipinski, J. M. Harrer, and R. L. Ramp

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Chapter 14, "Remote Handling of Radioactive Materials"

K. R. Ferguson, W. B. Doe, and R. C. Goertz

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Chapter 15, "Reactor Development Test Facilities"

L. W. Fromm, Jr.

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