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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

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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:

August 1962	ANL-6610
September 1962	ANL-6619
October 1962	ANL-6635
November 1962	ANL-6658
December 1962	ANL-6672
January 1963	ANL-6683

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I. BOILING WATER REACTORS

A. BORAX-V

1. Operations and Experiments

a. Reactor Operations. During February, BORAX-V was operated at pressure (600 psi) to check the performance of the boiling core and plant equipment. Operational testing of experimental equipment included calibrations of miniature in-core ion chambers, measurements of fuel-rod temperature and thermal conductivity, transfer function measurements, flux-wire irradiations, fuel-rod irradiations, xenon determination, process control and instrumentation testing, as well as determinations of reactor water level, carryover and carryunder, measurements of calorimetric steam quality, and operator training. A maximum power of 20.5 Mwt with all control rods fully withdrawn was reached. Steady-state reactor powers to 19.5 Mwt were maintained. The calculated total reactor power production to date is 65 Mwd.

The facility was also operated at zero power (under open-vessel conditions) to test the reactivity effects of a fueled rotating oscillator located in a reflector thimble adjacent to the core, substitute materials for control rods, and an in-core, step-function generator or reciprocating oscillator. This completed the experimental program on boiling core B-2.

Unloading of all 60 boiling fuel assemblies has been completed, and removal and inspection of control rods has started. Preparations are being made for installation of the core with central superheater.

b. Reactor Physics. Axial flux traverses were obtained by inserting reactor-grade zirconium wires into stainless steel flux wire pressure thimbles. Significant changes in the radial power shape were evident as power level increased. This shows that void formation near the center of the core causes a substantial reduction of thermal flux in that vicinity. Figure 1 gives the axial shapes obtained at given radial distances for 15-Mwt operation.

An irradiation of 15 fresh, 4.95% enriched fuel rods in selected positions in core B-2 for flux plotting was made at a power of 15 Mwt for 3 hr. The rods were then removed from the core; ten days later they read ~500 r/hr on contact. No counting has been done yet. The lead transport pig and the scanning pigs for fuel rods have been checked out and work satisfactorily.

The effect on reactivity of fission product poisons and fuel burn-up was measured. Forty-one and one-half hours after completion of 65 Mwd of power operation, a loss of reactivity of approximately 0.35% was observed.

Figure 1. Axial Flux Distributions at 15 Mwt - Boiling Core B-2 - BORAX-V

Following power operation, several changes were made in the core. The oscillator and assembly in core position 75 were removed and a fuel assembly installed. Six in-core flux wire thimbles were removed, and five miniature ion-chamber thimbles were replaced with fuel rods. Seven additional boron-stainless steel poison rods were installed. These changes resulted in a reactivity increase at room temperature of $\sim 0.7\%$.

A mockup fueled oscillator rod with a rotor, $2\frac{7}{8}$ in. in OD and 24 in. long, containing 126.5 g U^{235} over a 180° arc and with 0.040-in.-thick cadmium over the remaining 180° , was inserted into a reflector thimble and rotated to determine the total and differential worth in core B-2 at room temperature. The differential worth was determined to be $0.149\% \Delta k/k$ for 126.5 g of U^{235} , compared with the values of $0.110\% \Delta k/k$ and the $0.066\% \Delta k/k$ for 55 g and 16 g of U^{235} in similar oscillators (see Progress Report for November 1962, ANL-6658, p. 5). With the fueled portion of the oscillator turned in toward the core, the total worth of the oscillator was $0.122\% \Delta k/k$; with the poison turned toward the core, the total worth of the oscillator was $-0.031\% \Delta k/k$.

An experiment to determine the relative reactivity worths of various control materials was run on the clean core B-2 containing 40 poison rods and no thimbles or oscillator. The tests were run at room temperature with control rods banked at about 8.9 in. Samples of control materials were compared in a special fuel assembly which had one row of fuel rods removed to leave a vertical water slot through the core. Samples were 8 in. long, $3\frac{1}{2}$ in. wide, and $\frac{5}{16}$, $\frac{1}{4}$, or $\frac{3}{16}$ in. thick. The measured order of control effectiveness, based on $\frac{5}{16}$ -in.-thick samples, is listed in Table I. Analysis of results of this experiment is continuing.

Table I. Effectiveness of Control Materials

<u>Order of Control Effectiveness</u>	<u>Sample</u>
1	2 w/o boron (93% enriched in B^{10})-stainless steel
2	hafnium
3	5 w/o Cd, 15 w/o In, 80 w/o Ag, clad with 0.025-in. stainless steel
4	50 w/o Boral clad with 0.062-in. stainless steel (reference BORAX-V control-rod material)
5	2 w/o natural boron-stainless steel
6	cadmium

c. Reactor Transfer Functions.

(i) Measurements. Transfer-function measurements (to give values of $\Delta n/n_0 \Delta k$) by the sine-cosine cross-correlation technique were run

with core B-2 at 10 and 16.5 Mwt (see Progress Report for January 1963, ANL-6683, p. 2). The rotating poison oscillator in core position 75 was inserted to $\frac{1}{2}$ in. above the core bottom for these runs. To reduce the amplitude of the power oscillations near the resonant frequency from the ± 20 -30% noticed in these measurements of power transfer functions, the oscillator was subsequently withdrawn to the 12-in. position, and additional zero-power and 5, 10, and 15 Mwt transfer-function measurements were made with the oscillator in this position. The reduction in reactivity amplitude is shown in Table II. The frequency range studied was again 0.05 to 15.0 cps for the 5 Mwt run, but was increased to 0.008 to 15.0 cps for the 0, 10, and 15 Mwt runs.

The transfer-function results obtained with these two oscillator (axial) positions are being compared for effects due to nonlinearities associated with large, forced-power oscillations. These results are also shown in Table II.

Table II. Results of Transfer-function Measurements

Power Level, Mwt	Oscillator Position, in.	Deduced Reactivity Amplitude, cents	Flux Amplitude at Resonance Frequency, %	Gain of $\Delta k/n_0 \Delta k$ at Resonance Frequency	Resonance Frequency, cps
0 (489°F, 600 psig)	$\frac{1}{2}$ withdrawn	6.06	6.06	1.00(0 db)	-
	12 withdrawn	4.85	4.85	1.00(0 db)	-
5	$\frac{1}{2}$	-	-	-	-
	12	6.17*	8.4	1.36(2.68 db)	1.3
10	$\frac{1}{2}$	14.9*	19.0	1.56(3.86 db)	1.6
	12	7.27*	11.4	1.57(3.92 db)	1.7
15	$\frac{1}{2}$	15.8*	28.6	1.81(5.16 db)	2.0
	12	8.55*	15.0	1.76(4.92 db)	2.1

*Deduced by normalizing to the zero-power data at 15 cps.

In general, the deduced oscillator worth (differential reactivity amplitude) increases with increasing reactor power. There are at least two reasons for this: (1) the oscillator is increasingly "unshadowed" as the control-rod bank position rises with increasing power; (2) the increasing void fraction in the core increases the neutron diffusion length, thus increasing the oscillator rod worth (as well as all control rods).

To facilitate obtaining the following desired transfer functions, G , G_0H , and H , where

$$\frac{\Delta n}{n_0 \Delta k} = G = \frac{G_0}{1 + G_0 H} ,$$

an IBM-1620 code was devised to calculate the above three complex quantities vs. frequency directly from the results of the sine-cosine cross-correlator. The code is now checked out and operating satisfactorily.

d. Water Chemistry. Chemical monitoring of the reactor water during operating periods continued. Continuous measurements of resistivity, chloride concentration, and fission product activity were made, along with measurements of intermittent pH, concentrations of suspended solids, and gas analyses. Steam carryover tests were completed at 10 and 15 Mwt. The resistivities of the various streams remained at satisfactorily high levels.

During previous operating periods, it was discovered that a certain amount of backflow occurs in the reactor water demineralizer at the time it is shut down. This backflow, over a period of time, has been sufficient to separate partially the two resins in the mixed bed, with the result that the resin appears to be nearing exhaustion. By remixing the bed with air and using enough water to slurry the resin, the effluent water quality has been twice restored. The last time, the effluent quality dropped from 9 to about 2 megohm-cm. A mix restored the quality to 6 megohm-cm, which continued to the end of the operating period.

Chloride concentrations in the reactor water remained below 0.1 ppm, and for most of the operating period ran at 0.05 ppm or less. Chloride content of the condensate was 0.05 ppm or less.

The fission product monitor gave several high readings. The resin on the fission product monitor cleanup column and a particulate sample from the stack gas were gamma-scanned. Cs^{138} was identified in the resin and in the stack gas sample. The concentration of Cs^{138} was too low to indicate a major cladding rupture. The most likely explanation is that surface contamination was responsible for the Cs^{138} .

Intermittent pH determinations were made, indicating that the pH of the high-purity water was at, or very near, 7.

The concentrations of suspended solids are shown in Table III, along with the analysis of the main metallic constituents.

The high solids concentration of 0.23 ppm noted for the period 1/28-30/63 cannot be explained on the basis of the high conductivity caused by the sodium nitrate injected for the steam carryover and carryunder tests. The previous low value of 0.05 ppm was obtained just after the first two of these tests.

Table III. Concentrations of Suspended Solids

Sampling Period	Solids Concentration (ppm)	Analysis (w/o)				Point Sampled
		Fe	Cr	Ni	Al	
1/23-24/63	0.03	54.8	4.0	7.5	13.2	Reactor bottom
1/24-28/63	0.05	No analysis				Reactor bottom
1/28-30/63	0.23	9.1	1.6	1.7	37.9	Reactor bottom
1/30-2/1/63	0.01	29.0	1.8	5.1	40.0	Mid-vessel
2/6-8/63	0.08	9.3	1.0	1.8	40.0	Mid-vessel
2/8-13/63	0.07	23.0	0.8	4.3	43.0	Mid-vessel

The fuel elements, assembly boxes, feedwater sparger, steam probe, and other reactor vessel internals were visually examined for deposits and corrosion. All metal surfaces examined had a two-layer coat. The coating next to the surface of the metal was thin, hard, adherent, and grayish-brown in color. The outer coat was soft, nonadherent, extremely thin, and appeared to be similar to finely powdered rust. No pitting, corrosion, or heavy deposit was noted on X-8001 aluminum or stainless steel.

Seventeen determinations of total radiolytic gas were made over the entire two-month operating period at various powers and sample points. The gas was analyzed for its components in percent by volume. Two typical analyses are shown below for gas from condensed steam:

Sample	I	II
H ₂	56.6%	58.0%
O ₂	32.4%	30.4%
Inerts	11.0%	11.6%

The data from the steam-carryover tests are shown in Table IV. A movable steam probe in the steam dome was used for sampling.

Table IV. Results of Steam-carryover Tests

TEST I			TEST II		
Initial Conditions		Conditions after Sodium Nitrate Injection	Initial Conditions		Conditions after Sodium Nitrate Injection
Power (10 Mwt)		Power (10 Mwt)	Power, 15 Mwt		Power, 15 Mwt
Resistivity, 2.26 megohm-cm		Resistivity, 0.25 megohm-cm	Resistivity, 2.22 megohm-cm		Resistivity, 0.26 megohm-cm
pH, 6.2 (probably erroneous, due to high purity of water)		pH, 7.0	pH, 7.3 (may be erroneous, due to high water quality)		pH, 7.4
Probe Position from Reactor Bottom	Steam Quality % Moisture	Remarks	Probe Position from Reactor Bottom	Steam Quality % Moisture	Remarks
15 ft 9-3/4 in.	0	Probe fully withdrawn (vessel height, 16 ft)	15 ft 9-3/4 in.	0	Probe fully withdrawn
15 ft 1-1/4 in.	0		15 ft 1-1/4 in.	0	Level of steam collector opening
15 ft 1/4 in.	0		15 ft 1/4 in.	0	
14 ft 8-1/4 in.	0	Probe fully inserted	14 ft 8-1/4 in.	0	Probe fully inserted
14 ft 3-1/4 in.	0.07		14 ft 3-1/4 in.	8.5	
13 ft 10-1/4 in.	78.3		13 ft 10-1/4 in.	93.4	
13 ft 6-1/4 in.	92.1		13 ft 6-1/4 in.	95.9	
13 ft 1-1/4 in.	94.2		13 ft 1-1/4 in.	97.6	
12 ft 8-1/4 in.	94.4		12 ft 8-1/4 in.	96.8	
12 ft 3-1/4 in.	93.0		12 ft 3-1/4 in.	97.2	
11 ft 10-1/4 in.	94.0		11 ft 10-1/4 in.	96.8	

Steam quality in the steam line, determined by means of a throttling calorimeter, was found to exceed 99.5% at all powers measured to 15 Mwt.

e. In-vessel Instrumentation. Initial analysis of some temperature and flux measurements taken during steady-state and transient operation of the reactor indicates the following:

(i) Fuel Temperatures. Indicated temperatures, as a function of time at constant power, showed an increase over a two-month period of operation for four of the five fuel-center thermocouples. Scram tests have been run, but not yet analyzed, to try to separate the effects of thermocouple aging on the above measurements (thermocouple-calibration shift and thermal conductivity of fuel could produce the same effect at steady-state conditions). The maximum fuel temperature of 1810°F was recorded at 19Mwt in fuel rod 18, core position 46. This location gave the hottest temperature reported previously (see Progress Report for January 1963, ANL-6683, p. 5), although the indicated temperature now is considerably higher than that reported for a higher power (i.e., 1675°F at 20.5 Mwt).

Table V shows measured temperatures as a function of power and fuel age. Fuel thermocouple locations are shown in Figure 2. All thermocouples are 10 in. up from fuel bottom except T/C-4, which is 9 in.

Table V. Fuel Temperature as a Function of Power

<u>Date</u>	<u>Indicated Temperature (°F)</u>					<u>Power (Mwt)</u>
	<u>T/C-2</u>	<u>T/C-3</u>	<u>T/C-4</u>	<u>T/C-5</u>	<u>T/C-6</u>	
12/20/62	810	890	790	790	780	5
1/8/63	-*	962	840	840	840	5
1/29/63	-	1420	940	1280	1260	5
2/7/63	820	-	970	1250	1260	5
1/8/63	-	1195	1100	1080	1110	10
1/17/63	-	1258	1150	1170	1170	10
1/18/63	-	1256	1140	1190	1170	10
1/23/63	1185	1540	1160	1320	1240	10
2/6/63	1150	-	1360	1560	1560	10
2/6/63	1310	-	1550	1635	1670	15
1/29/63	-	1810	1570	1710	1700	19
1/22/63	1480	1615	1430	1490	1490	20
2/4/63	-	1270	1500	-	1640	20
1/22/63	1520	1690	1480	1570	1560	22

*Thermocouple in use for other tests.

Figure 2. Fuel Thermocouple Locations - Core Position 46, Core B-2 - BORAX-V

(ii) Reactor-vessel-material Specimen Temperatures. Measurement of specimen temperatures has not led to the detection of any measurable temperature rise above water temperature during power operation up to 20.5 Mwt.

(iii) Neutron-flux Measurements. Neutron-flux measurements were obtained with a standard (fixed location) and dual-chamber unit during power operation. The dual-chamber unit utilized one cadmium-shielded chamber packaged with an unshielded chamber. Traverses were made with the latter unit in two in-vessel thimbles at powers of 5, 10, and 15 Mwt and data obtained for cadmium ratio measurements. There was difficulty with data from the cadmium-shielded chamber; an exponential buildup and decay of a reverse signal current was experienced in this chamber. Time for full buildup or decay was found to be approximately 8 min.

Zirconium wires were irradiated for backup in each of the above cases, and flux wire plots are in rough agreement with ion chamber data.

(iv) Water Level Measurements. Using the movable in-vessel steam probe and the sodium-tracer technique, reactor water level was measured as 13 ft 1 in. for a power of 1 Mwt, while reactor vessel water level recorder LR-1 was recording 14 ft 2 in. and the reactor vessel water level sight-glass LG-1 reading was 12 ft 8 in.

Another test was made at 600 psig, 490°F, and zero power, to check the comparative readings of LR-1 and the sight-glass. The test was run by closing the lowest vessel connection to the level standpipe (three connections join the standpipe to the vessel) and draining the vessel to an elevation corresponding to the next lowest standpipe connection. This elevation was indicated, during draining, when one of the standpipe-connected instruments stopped changing its indicated level. This first occurred for the sight-glass LG-1, which read 11 ft 5 $\frac{1}{2}$ in. At the same time, the level recorder LR-1 read 13 ft 0 in., with the true elevation being 10 ft 4 $\frac{3}{4}$ in.

A design study to provide circuits for correcting the action of LR-1 is being made.

2. Procurement and Fabrication

a. Superheater Fuel. All fuel for the central superheater has been received at the reactor site. This includes 13 standard, three instrumented, and one cutaway display assembly containing depleted UO_2 .

Fabrication and inspection of 17 standard and two instrumented peripheral superheater fuel assemblies has been completed. This completes the fabrication of all the reference design superheater fuel.

b. Type 406 Stainless Steel-clad Superheat Fuel. Atomics International has fabricated six depleted Type 406 fuel plates which they will evaluate prior to beginning work on enriched fuel plates of this type.

Blended UO_2 and Type 406 stainless steel materials for five fuel plate cores were cold pressed at Armour Research Foundation at 50-51 tsi and the pressed cores were vacuum heat treated at 200-250°C for 11 hr. Picture frames and cover plates have been descaled and/or electropolished and compacts for rolling are being prepared.

Type 406 stainless steel dummy fuel plates, side strips, and spacer wires have been descaled. Solutions for electroplating with nickel are ready and procedures for plating these descaled subassembly components are being developed.

c. Advanced Superheat Fuel Elements. Superheat fuel elements of an advanced concept have been proposed for BORAX-V. Details of the design were reported in the Progress Report for December 1962, ANL-6672, p. 6.

Dies are being prepared for fabrication of 0.355-in.- and 0.321-in.- diameter UO_2 balls, and for forming 0.375-in.-OD x 0.010-in.-wall, hemispherical cladding sections. Swaging is being considered as a method for reduction of the tubing around 0.321-in. UO_2 balls or similarly forming tubing filled with vibrator compacted UO_2 powder. Another method considered involves isostatic pressing cladding tubing around UO_2 balls.

Although the cladding material has not been definitely chosen, both welded and seamless Type 406 stainless steel tubing and material for extrusion have been ordered for development purposes.

II. LIQUID-METAL-COOLED REACTORS

A. General Research and Development

1. Physics

a. ZPR-III, Assembly 42. Experiments with Assembly 42, a two-zone reactor (see Progress Report for January 1963, ANL-6683, p. 7) having a simulated uranium-carbide core and a uranium-carbon-steel annular driver, continued this month.

i. Fission Traverses. Axial fission traverses were completed this month. Two opposing drawers at the center of the reactor were modified to accept a $\frac{1}{2}$ -in.-diameter thimble to make the traverses. Small counters were moved remotely in this thimble. The relative fission rate as a function of axial position was measured for U^{235} , U^{238} , Pu^{239} , and U^{234} , as well as the reaction rate for B^{10} from an enriched BF_3 counter.

Radial fission traverses were made through the core center by modifying a vertical column of drawers in Half No. 2 to accept a $\frac{1}{2}$ -in.-diameter traverse thimble. The position of this thimble, when in place, is approximately $\frac{1}{2}$ in. from the core face. The relative fission rate as a function of radial position was measured for U^{235} , U^{238} , Pu^{239} , and U^{234} , as well as the reaction rate for B^{10} from an enriched BF_3 counter.

ii. Nuclear Track Emulsions. The nuclear track emulsions exposed last month at integrated fluxes of 1×10^8 , 5×10^8 , and 1×10^9 nvt were found to be fogged and were unacceptable for counting. A new set of plates were exposed at increased integrated fluxes of 1×10^9 , 3×10^9 , and 6×10^9 nvt, and sent to Northwestern University for analyses. Although the exposures were adequate, several plates were damaged in transit. Consequently, a third exposure may be necessary.

iii. Worth of Core Drawers. The reactivity worths of core drawers at three different radial positions were measured, relative to void, by the technique of subcritical multiplication determination. The results are given in Table VI.

Table VI. Reactivity Worth vs. Position

<u>Position</u>	<u>Material</u>	<u>Worth (lh/drawer)</u>
Center	Core	106.7
Core Edge	Core	83.8
Driver	Driver	130.3

iv. Rossi-alpha Measurements. Since this reactor consisted of two zones differing greatly in composition, three different arrangements of four BF_3 counters were used to determine if the different lifetimes of each region could be measured. There was no noticeable difference between the values of the driver and core alphas, however. The single observed value of alpha appears to agree with the weighted averages of the alphas for Assemblies 34 and 17, corrected for slight differences in composition. The observed value of alpha, $3.70 \times 10^4 \text{ sec}^{-1}$, and the assumed delayed-neutron fraction of 0.0072 lead to an average lifetime of $19.5 \times 10^{-8} \text{ sec}$.

v. Central Reactivity Coefficient Measurements. The reactivity changes due to insertion of various fissile and nonfissile materials into the center of the core have been measured. These measurements were made by creating a $\frac{1}{4} \times 2 \times 2$ -in. void for fissile samples and a $1 \times 2 \times 2$ -in. void for nonfissile samples at the front of two opposing central drawers. The fissile samples were held rigidly in place by the use of full-density aluminum inserts. For these measurements, the reference core was run with reduced-density aluminum in the void region so there would be no net change in aluminum mass. The preliminary results of these experiments are given in Table VII.

Table VII. Central Reactivity Coefficients

Sample	Sample Mass (kg)	Worth (lh/kg)	Sample	Sample Mass (kg)	Worth (lh/kg)
U^{235}	0.268	$117 \pm 2.0^{(a)}$	Al	0.35	0.2 ± 0.5
Pu	0.186	$187 \pm 2.5^{(b)}$	Ph-I	0.232	$-15.3 \pm 2.5^{(c, d)}$
U^{233}	0.221	$223 \pm 2.5^{(a)}$	Ph-II	0.210	$-14.5 \pm 2.5^{(c, d)}$
U^{238}	2.434	$-8.0 \pm 0.25^{(a)}$	Nb	0.489	$-16.6 \pm 1.0^{(c)}$
Th	1.488	-13.0 ± 0.3	Mo	1.279	-10.3 ± 0.5
SS	1.016	-1.7 ± 0.5	Hg	1.383	$-5.4 \pm 0.5^{(c)}$
Fe	1.028	-1.6 ± 0.5	Al_2O_3	0.355	8.3 ± 1.5
Ni	1.15	-2.5 ± 0.5	Y	0.582	-0.6 ± 1.0
Cr	0.439	$-0.7 \pm 1.0^{(c)}$	Li	0.0549	$-97.6 \pm 10.0^{(c)}$
Zr	0.846	-1.3 ± 0.5	Be	0.24	83.1 ± 2
Na	0.091	$10.3 \pm 5.0^{(c)}$	B_4^{10}C	0.0166	$-1437 \pm 30^{(e)}$
Pb	1.474	-0.25 ± 0.3	Ta	1.004	$-30.2 \pm 0.5^{(c)}$
C	0.196	35.5 ± 2.5	Ag	0.683	-50.4 ± 0.8
Bi	1.27	-0.07 ± 0.4			

^(a)Reactivity of principal isotope only.

^(b)Reactivity of total sample containing 95% Pu^{239} .

^(c)Corrected for effect of stainless steel container.

^(d)Ph-I and Ph-II are mixtures of metals and metal oxides used to simulate fission products.

^(e)The boron carbide sample contains 69.34% boron enriched to 90.7 atom percent in the isotope B^{10} .

2. Fast Reactor Fuels

a. Corrosion of Fuel-cladding Materials. Three vanadium-base alloys which have been tested in flowing sodium ($\sim 2 \text{ fps}$) for 504 hr at 650°C at a reported Na_2O concentration of 1-5 ppm have been sectioned, metallographically prepared and photographed, and microhardness measurements across the sections have been taken. The alloys are: (1) V-10 Ti-3 Nb; (2) V-10 Ti-3 Ta; and (3) V-5 Ti-20 Nb, where the compositions given are in weight percent.

All the alloys tested in sodium showed a darkly etched band near the surface of the material, which was followed by a white layer at the surface. Electron-diffraction techniques are being employed to try to determine the structure of this surface layer in alloy (3). The structure of the as-received specimens was uniform throughout.

Microhardness measurements showed the edge hardness of all the test specimens to be much greater than the center hardness. The as-received specimens showed a constant hardness across their transverse sections. In all cases, the center hardness of the as-received specimens was higher than the center hardness of the respective sodium-tested specimens. This is thought to be a result of a partial softening at the temperature of the test. Specimens of as-received material were vacuum annealed for 23 hr at 650°C. All the specimens softened and approached the hardness of the respective sodium-tested specimens. The data are given in Table VIII.

Table VIII. Results of Hardness Tests

<u>Alloy</u>	<u>Edge Hardness (T)</u>	<u>Center Hardness (T)</u>	<u>Center Hardness (C)</u>	<u>Center Hardness (V.A.)</u>
1	650	190	240	200
2	450	195	240	200
3	680	235	275	245

(T) Tested in sodium

(C) As-received

(V.A.) Vacuum Annealed Control

As-received and test specimens are being analyzed for oxygen content.

The vanadium-base alloy samples were too small to be tested for ductility in the free-bend-testing device described in previous reports. However, the hardness values indicate that, as in the case of Nb-4.33w/o Zr and Nb-1.84 w/o Cr, these alloys would also have been embrittled after exposure to sodium.

Attention is called to the statement in the section of the December, 1962 report* dealing with FARET, which discussed the usefulness of niobium- and vanadium-base alloys for fuel-cladding material. There is no experimental evidence which indicates that presently available niobium and vanadium alloys are compatible with sodium of the quality likely to be found in a practical reactor system, i.e., sodium containing 20-100 ppm oxygen. High-temperature mechanical properties and compatibility with carbide fuels are also in doubt.

*ANL-6672, p. 21

b. Irradiation Tests. Instrumented capsules are irradiated in the CP-5 reactor as part of a program for the development of fuel elements for fast reactors. Currently, irradiated capsules CP-21, -24, -25, and -26 are waiting to be disassembled and for the contents to be examined in the hot cells. Capsules CP-27, -28, -29 and -30 are in the reactor and capsule CP-31 is ready to go in.

All of the capsules, except CP-26, -29, and -31, were made of Zircaloy-2 duplex tubing. The three exceptions were made from Type 304 stainless steel duplex tubing. Heaters and thermocouples were incorporated in each capsule. NaK was used as a heat transfer medium in all capsules except CP-26, in which sodium was employed. With the exception of CP-24 and -26, each capsule contains six fuel specimens in two tiers of three. CP-24 contains one PuC specimen and CP-26 contains 16 specimens of tubing made from alloys which might serve for fuel containment. The fuel in the specimens in capsules CP-21, -25, -27, -28, and -29 is U-20 Pu-10 Fs. CP-30 contains specimens with fuel of the following compositions: U-10 Pu, U-15 Pu, U-10 Pu-10 Fz, and U-15 Pu-10 Fz. The fuel in capsule CP-31 is beta treated uranium, 20% enriched. In general, the cross-section geometry of the fueled specimens is the same as for the standard EBR-II rod. The length of the fuel section (usually 2 in.) and the void space over the fuel were in proportion to a standard EBR-II fuel rod, again with some exceptions. The containment tubing was 0.175-in. O.D. with a 9-mil wall in all cases, including the tube specimens. Most of the fuel specimens have a sodium bond to the cladding.

In capsule CP-21, removed with an accumulated burnup of about 1.95 a/o, three specimens were clad with Nb-1 w/o Zr alloy and three with Ta-1 w/o W alloy; irradiation temperature at the center of the fuel was 580°C. Capsule CP-24 attained a burnup of about 5 a/o. Capsule CP-25, removed after an estimated burnup of 2.35 a/o, contained two specimens with Nb-1 w/o Zr cladding, two with Ta-0.1 w/o W cladding, and two with Nb-33 w/o Ta-1 w/o Zr; the maximum central fuel temperature was about 620°C. Capsule CP-26 was removed after 471-Mwd exposure because of various malfunctions of equipment.

The purpose of the experiment with capsule CP-27 is to determine if the release of buildup gas pressure will extend specimen life. The experiment with capsule CP-31 is being made to determine the irradiation behavior of vented metallic fuels.

3. Preparations for ZPR-VI and ZPR-IX Operations

The ZPR-VI facility contains a fast critical reactor assembly designed primarily for, but not limited to, the investigation of the sodium void and prompt Doppler coefficients in large dilute systems. The non-nuclear testing of the ZPR-VI facility was essentially completed several

months ago; nuclear testing is being held in abeyance pending approval by the AEC of the Safety Analysis Report.

Special equipment for making various types of measurements in the ZPR-VI system is being assembled. A mechanical oscillator designed for measuring Doppler effect has been installed and is being tested. The equipment to be used for making Rossi-alpha measurements is being tested with the Argonne Thermal Source Reactor. The equipment to be used for making interim reactor period measurements has been assembled, and its use in conjunction with a simple analog computer to provide direct results is being investigated.

The emergency air system for purging the cells containing both the ZPR-VI and ZPR-IX systems has been tested and results were found to compare favorably with the design specifications. A conveyor to move loaded fuel drawers between the work area and the cell has been installed and satisfactorily tested.

It is initially planned to use the ZPR-IX facility in support of the nuclear rocket reactor study. Critical experiments are required initially to define the basic physics data pertinent to the design of proposed systems. Initially clean critical studies would be made on moderate-sized assemblies containing tungsten and U^{235} in the core, and heavy metal and beryllium in the reflector. As the studies progress, it would be expected that cores having a volume up to 800 liters and a void fraction as high as two-thirds would be investigated.

It is estimated that the ZPR-IX facility will be ready for nuclear operation in late spring. The final Safety Analysis Report is also scheduled to be completed about that time.

The principal components yet to be installed in the ZPR-IX facility are the aluminum matrix tubes, the aluminum fuel drawers, and most of the control and safety-rod drive units. An additional six weeks is required to complete bundling and welding of the last twenty-five aluminum matrix tubes. Approximately 40% of the required aluminum drawers for the matrix assembly have been fabricated. Assembly and testing of the balance of control and safety rod drive units are currently underway.

B. EBR-I, Mark IV

Fission destruction patterns of Pu^{239} throughout the EBR-I, Mark IV core have been established through the activation of aluminum-clad plutonium foils at low power (1 to 5 kw). In a similar manner, fission destruction patterns of U^{235} have been established throughout the upper and lower axial blankets, the light radial blanket, and the heavy outer uranium

cup by means of activating metallic uranium foils enriched in U^{235} . Although the analysis of various details is still incomplete, results appear to be satisfactory.

Traverses with metallic foils of depleted uranium throughout the various blankets have also been completed. However, attempts to establish the Pu^{239} production patterns from these traverses have not yet been successful. The difficulty encountered originates apparently from the inability to resolve a pure Np^{239} component from the depleted monitor foils irradiated in a reasonably well-thermalized graphite position. This is surprising, since no difficulty has been encountered with the separation of a pure Np^{239} component from the substantial fission product background associated with depleted foils irradiated in the very hard neutron spectrum at the core center. Superficially, it would seem that any method capable of separating a Np^{239} component from a depleted foil irradiated in a fast flux would almost certainly succeed for a similar foil irradiated in a highly energy-degraded neutron spectrum. Experiments intended to resolve this perplexing discrepancy are in progress.

C. EBR-II

1. Reactor Plant

The final pre-operational test of the fuel-handling system for the reactor was completed; the primary tank received a final cleanup, and the overall system was rigorously inspected. The M-2 primary sodium pump was reinstalled in its nozzle, which had been previously used for access into the primary tank. An additional heater was inserted temporarily into the primary tank through a nozzle near the heat exchanger.

The primary tank cover was heated to about 250°F and the rotating shield plug seals were filled with low-melting Cerrotru alloy, composed of 58% bismuth and 42% tin. The large plug seal trough (see Monthly Report for August 1962, ANL-6610, p. 24, Fig. 10) required approximately 2100 lb, and the small seal trough required approximately 1300 lb, of alloy. After filling the troughs, the heater control system and the new seal system (which provides an argon atmosphere above the Cerrotru) were tested and adjusted. The rotation and operation of both shield plugs were given a final test. The argon leak rate of the mechanical seals above the Cerrotru troughs was expectedly excessive due to large clearances existing at the low (200°F) structural temperature. With an argon purge rate of 120 cu ft/hr for the large plug and 80 cu ft/hr for the small plug, an oxygen concentration of less than 2.5% was obtained. This was considered satisfactory under these conditions. Nitrogen purging of the tank continued throughout this period and until sodium filling started. About 90,000 cu ft of nitrogen was expended during the purge to reduce the oxygen concentration to 0.56%.

After the rotating shield plug seals were filled and various other openings were sealed off, the primary tank cover and the associated piping systems were surveyed for leaks by a helium mass spectrometer. A differential pressure of 2 to 6 in. of H_2O was used; no leakage of gas from the primary tank was detected.

The primary tank was filled with 88,000 gal of sodium which were pumped at 275°F by means of pressurized (15 psi) nitrogen through filters from nine storage tank cars located outside the reactor plant. Initially, the maximum flow rate was about 60 gpm. During the unloading of the fifth car, the in-line micrometallic filter (20- μ pores) started to plug and the flow rate was reduced to about 30 gpm during transfer from the sixth car. Filling, which started on February 18, continued at the rate of approximately one railroad tank car per day and was completed on February 26. Preliminary visual examination by means of a periscope indicated that the sodium surface was clean.

An extra half tank car (5,000 gal) is being unloaded into drums for reserve storage. Sodium circulation through temporary cold traps has been started.

2. Sodium Boiler Plant and Power Plant

After the storage tank was filled, recirculation of sodium was established through the cold trap (isothermally). The steam system, secondary sodium system piping, and the storage tank were brought to 350°F in preparation for filling the secondary system with sodium from its drain tank. However, the operation was temporarily postponed because of the following difficulties: (1) a drain-valve heater failed, making the valve inoperable; (2) a malfunction occurred in the voltage-control circuit of the main secondary pump; and (3) while work was in progress for correction of Item 2, an electrical shortcircuit in the main power leads to the secondary pump caused some insulation damage. In view of these delay factors and the readiness of the primary system for sodium filling, it was decided to shut down and cool the steam system and secondary system and keep the storage tank and purification system hot.

Circulation has been maintained isothermally at 350°F through the purification system. Bulk sodium samples have been taken and submitted for chemical analysis, and operating tests indicated a plugging temperature of about 260°F.

Experience obtained during the heat-up to 350°F indicated that to achieve maximum circulation and an even heating rate, a so-called solid system, with the boiler steam drum, superheaters, and associated piping completely filled with water, is necessary. The most serious difficulties were encountered with the blowdown system and the No. 2 deaerating feedwater heater.

It was deduced during this phase of operation that the difficulty encountered in placing the low-pressure flash tank in service was caused by flashing in the blowdown cooler. This occurs because the cooler is at a slightly higher level than the flash tank. Flashing of saturated liquid under this condition is inevitable, because of reduced pressure at the higher elevation. Since relocation of the tank to the next lower deck is impractical at this time, the injection of cool condensate into the line between the flash tank and the blowdown cooler in order to effect subcooling is being provided for.

The No. 2 heater was opened and inspected in an attempt to ascertain the cause of the water hammer which plagued operation during the entire heating cycle. This inspection revealed that condensate spray tubes had been installed incorrectly. The tubes have been reinstalled with the proper orientation, and new spray tubes of modified design for better operation at low loads have been ordered.

The steam system is being maintained in standby condition, flooded with water containing a rust inhibitor.

During shutdown, minor systems modifications are being performed as pointed out by limited operating experience to date.

3. Fuel Cycle Facility

Operation of the Argon Cell with an atmosphere primarily consisting of nitrogen (see Progress Report for January 1963, ANL-6683, p. 13) was continued. Tests to determine rate of air inleakage to the 60,000 ft³ volume under nonoperating conditions by measuring the rate of increase in oxygen concentration at a pressure differential of 4 in. of water showed a very acceptable leak rate of less than 0.005 cu ft/min. The oxygen-removal system operated effectively to reach levels of the order of only a few ppm oxygen, although continued difficulty was encountered with the analytic instrumentation. This appeared to be due in part to leaks and plugs in the sampling manifold. A number of transfers into and out of the cell, by means of both large and small transfer locks, were accomplished without marked effects on either the composition of the cell atmosphere or on the pressure-control system.

In making one of the transfers, the clamping screw on the large lock cover failed. The screw was successfully replaced remotely. When the gasket on the small lock cover stuck to the lid, the gasket dropped on the cell floor and was replaced remotely, although with considerable difficulty. Small permanent magnets have been obtained to help hold the gasket on the lock face.

In-cell transfers included all components for the decanning machine, and remote assembly of this unit is underway. The two compressors for the cell-atmosphere pneumatic-actuator system were started up with some minor difficulties, and it was noted that the loss of cell gas to the suspect exhaust system appeared to be excessive (of the order of 1 cu ft/min).

Equipment for extruding, measuring, and loading sodium into fuel-element tubes has been placed in a glove box. The glove box has been fitted with windows and ports, and its atmosphere-purification system has been received.

Design work is now completed on all major items of process machinery. The fuel-element gaging and tensioning-tester drawings were completed. Loading fixtures and storage racks for fuel-element components, furnace crucible, pallets, and special tools were also designed. Drawings for the fuel-element-assembly machine were checked and coordinated with the changes requested by the vendor. As-built drawings were completed for the fuel-pin-process machines.

The fuel-rod-welding machine was set up and operated. Revision of the restrainer delivery chute was required and locking devices were installed on all fasteners to prevent loosening during operation.

A clutch of ANL design has been successfully tested for use with the drive motors which move the shielding window shutters (see Progress Report for January 1963, ANL-6683, p. 13). The use of a clutch will eliminate the chatter in the shutter drives which occurs at the end of shutter travel because of the high stalling torque of the drive motor. Clutches have been ordered for all shutter drive units.

A large transfer lock (of 6-ft diameter), which is mounted vertically in the floor of the Argon Cell, is used for moving large items between the Argon Cell and the Air Cell. A new laminated rubber and steel gasket has been designed for sealing the top closure (a counterbalanced, hinged lid) of this lock and is being evaluated as a replacement for the lead-faced steel gasket now used. Both gaskets have a diameter of about $6\frac{1}{2}$ ft, and it is necessary, therefore, to deform the gaskets when bringing them through the large transfer lock. The mechanical problems associated with equipment needed to control the deformation of the lead-faced steel gasket are believed to be greater than those with the newly designed gasket. The new gasket can be readily folded since it consists of a one-piece flat rubber ring cemented to a steel ring which is in two segments.

A portable lighting unit, mirror, and stand have been built to provide brilliant lighting for local areas within the Air Cell and the

Argon Cell. The light source is a 500-w Quartzline* lamp which has a life expectancy of about 2000 hr. Remote portability, adjustment, and replacement of parts have been provided in the design.

An off-gas filter unit has been designed for use in a vacuum system and will be fabricated by a commercial filter manufacturer. The filter will be made of radiation-resistant components, which will make it suitable for use with the melt-refining furnace, the skull-oxidation equipment, and the skull-oxide-processing equipment. The filter unit will utilize glass filter media and will contain charcoal for adsorbing fission product iodine which is released during the melt-refining operation. The filters will be attached to equipment vacuum lines by means of low-melting alloy seals. After removing a filter from a vacuum line, it will be capped to prevent spread of radioactive particulate material.

The skull-reclamation process provides for the oxidation of the skull to transform it to a powder for ease of removal of the material from the melt-refining crucible. A furnace for the oxidation step has been constructed and tested at Argonne (see Progress Report for December 1962, ANL-6672, p. 18). Associated with the furnace is an oxygen-feed-control system which regulates the rate of the oxidation reaction. Pressure-time data obtained by means of a pressure recorder for the oxidation furnace may be used to calculate (1) the instantaneous oxidation rate, (2) the concentration of oxygen in the furnace, (3) the total amount of oxygen consumed by the skull, and (4) the amount of oxygen discharged from the furnace. A preliminary run was made to test the oxygen-feed control system during the oxidation of a 550-gm skull contained in a zirconia crucible. The skull was oxidized to a fine powder. A maximum rate of 1.5 gm uranium/min was achieved in the oxidation of the uranium in the skull material to uranium oxide (U_3O_8). About 70 percent of the oxygen fed to the furnace was utilized in the oxidation reaction.

A dumper which will separate skull oxide from the zirconia crucible has been tested after being modified to eliminate minor mechanical operating interferences. Its operation is now considered satisfactory.

4. Fuel Development

a. Fuel-jacket Development for Core II. In addition to the three niobium alloys (Nb-5 w/o Zr, Nb-33 w/o Ta-1 w/o Zr, and Nb-10 w/o W-10 w/o Ta) mentioned in the Progress Report for January, ANL-6683, p. 13, another alloy, NB-10 w/o Ti-5 w/o Zr (D-36), is also being fabricated into 0.156-in.-I.D. x 0.015-in.-wall tubing.

b. Properties of Uranium-Plutonium-Fizzium. The properties of U-Pu-Fz alloys (see Progress Reports for November, 1962 and January, 1963) are being studied in order to determine the optimum composition and heat treatment for the Core II loading of EBR-II.

*An incandescent, filament-type, quartz-envelope lamp which contains added iodine. The iodine prevents the tungsten that is evaporated from the filament from accumulating on the lamp walls. The lamp is a product of the General Electric Company.

Thermal-cycling tests (see Progress Report for January 1963, ANL-6683, p. 15) resulted in drastic elongation and extreme porosity of a U-10 w/o Pu alloy, but no appreciable changes of U-10 w/o Pu-10 w/o Fz and U-15 w/o Pu-10 w/o Fz alloys. The latter transform much more slowly than do the binary alloys. These alloys were therefore subjected to thermal cycling with much more time at temperature to determine their stability to such treatment.

Thermal-cycling specimens, 1 in. in length and 0.143 in. in diameter, were submerged in NaK and sealed in a stainless capsule under $\frac{1}{2}$ atmos of helium. They were cycled 100 times between 450°C and 700°C, held for one hour at 700°C and 4 hr at 450°C. It took approximately 15 min to heat and cool the specimens. The U-10 w/o Pu alloy again showed very large changes: a 29% decrease in density and a 25% increase in length. The surface of the pin was very rough and swelled appreciably. The U-10 w/o Pu-10 w/o Fz and U-15 w/o Pu-10 w/o Fz alloys again showed little change as the result of thermal cycling.

The transformation kinetics of the U-10 w/o Pu and U-10 w/o Pu-10 w/o Fz alloys are being studied. Metallographic and hardness studies have indicated that homogenization is complete in the U-10 w/o Pu alloy after one day at 900°C. Because of a lower solidus, the equilibration for the U-10 w/o Pu-10 w/o Fz alloy was made at 850°C. Hardness and metallography indicate very little change after heat treating for two days in the complex alloy.

The brittleness of the U-Pu-Fs and U-Pu-Fz alloys have been studied with a Hounsfield Impact Tester.¹ The effect of alloy composition and of heat treatment gave only slight differences in impact strength. In all cases an extremely low value of about 1 ft-lb was obtained, whereas cold-worked low-carbon steel gives a value of 20 ft-lb in this equipment. A bend test has been found to be more effective in comparing the brittleness of these materials. The bend test showed the injection-cast U-10 w/o Pu-10 w/o Fz, homogenized at 800°C for seven days and held at 650°C for two days to stabilize high-temperature phases, had twice the bend strength of as-cast U-10 w/o Pu-10 w/o Fz. Homogenization and heat treatment for stabilization of low-temperature phases at 450°C appeared to result in little change in brittleness compared with the as-cast condition.

c. Uranium-Molybdenum Alloys. Further attempts (see Progress Report for January 1963, ANL-6683, p. 31) have been made to roll 1-in.-diameter U-14 w/o Mo alloy. The slugs were clad in Zircaloy and heated in molten salt.

¹Annual Report for Metallurgy Division for 1961, ANL-6516, pp. 403-409.

Light reductions of 3 to 4% per pass were tried at 1115-1150°C. Although the alloy seems to have withstood a total reduction of 6.5%, the material failed at about 19% total reduction. Heavy reductions of 11-13% per pass at the same temperature resulted in cracking at 24% total reduction. These heavy reductions at 1100°C, however, showed no visible evidence of failure.

5. Process Development

a. Skull-reclamation Process. The skull-reclamation process is being developed as an auxiliary to melt refining for processing EBR-II fuel. The skull-reclamation process has two purposes: (1) recovery of fissionable materials from melt-refining residues (skull material) and (2) separation of fission products from uranium. Demonstration runs are being performed in an argon-atmosphere glove box to determine permissible ranges of process conditions and to disclose possible difficulties in the skull-reclamation process. Two runs have been performed recently, each using 150 gm of skull oxide from a batch containing 10 percent of fine zirconia crucible fragments. After extraction of noble metals from a molten flux-skull oxide mixture at 800°C and reduction of uranium oxide to the metal at 800°C, material-balance calculations showed that only 50 and 35 percent of the zirconium oxide in the original flux-skull oxide mixture had been reduced to metal. Since other current work indicates that essentially no volatile zirconium compounds form in the process flux at a temperature (720°C) near the noble metal extraction and oxide reduction temperature (800°C), the unreduced zirconium oxide is apparently removed from the process stream in the waste flux. A following step consisted of precipitation of a uranium-zinc intermetallic compound from 5.5 weight percent magnesium-zinc solution at 525°C. Some of the zirconium precipitated and was carried along with the uranium, apparently because it was present at concentrations in excess of its solubility in the metal solution.

Over the entire runs, zirconium removals were 85 and 87 percent of the total zirconium introduced. These removals are considered to be adequate for normal skull oxides from the first few reactor cores if the oxides contain no more than 3 w/o zirconia fragments.

An allowable zirconia fragment level in the skull oxide of 0.8 w/o was previously calculated on the basis of zirconium solubility in 5 w/o magnesium-zinc at 500°C. The higher allowable concentration of 3 w/o zirconia fragments given in the preceding paragraph was calculated assuming (1) incomplete reduction of the zirconium oxide contained in the skull oxide and (2) intermetallic compound precipitation at 525°C,* rather than at 500°C, which improves zirconium solubility by a factor of about 1.8.

*The process temperature for intermetallic precipitation has not yet been chosen.

Satisfactory removal of fission products was demonstrated in the two demonstration runs. It was found, however, that agitation variables influenced removals in the noble-metal-extraction step. In a run with the agitator positioned $\frac{1}{4}$ in. below the flux-metal interface, ruthenium extraction was 93 percent, whereas in a similar run with the agitator $1\frac{1}{2}$ in. below the interface, ruthenium removal was 55 percent. Adequate uranium recovery was demonstrated in both runs. In the first run, uranium present in the reduction heel solution was treated as being lost and the calculated uranium yield was 85 percent; in the second run, processing of the skull oxide charge was begun in a crucible containing the heel from the preceding run, and uranium recovery was 95 percent. Better uranium recoveries are anticipated in large-scale runs than those obtained in the present runs.

b. Material and Equipment Evaluation. Although pressed-and-sintered tungsten crucibles have been shown to be satisfactory for the noble-metal-extraction and reduction steps of the skull-reclamation process and thixotropically cast beryllia crucibles have shown promise of being satisfactory for the precipitation and retorting steps, fabrication of crucibles from each of these materials is expensive, and the size of such crucibles may be limited. A possible method of overcoming both of these problems is by plasma spraying of materials having good corrosion resistance on crucibles fabricated from other, less-expensive materials which can be fabricated into large shapes. By proper use of available techniques, it may be possible to obtain adherent coatings even with coating and crucible materials having different coefficients of thermal expansion.

In order to survey the applicability of the plasma spraying to the coating of crucibles, samples of possible substrate materials, approximately 3 in.² in area, were plasma-spray-coated with tungsten, beryllium oxide, tantalum monoboride, or zirconium diboride at Western Gear Corporation, Lynwood, California, by various techniques. The sprayed samples were tested by thermal cycling between 300 and 800°C for a total of twelve times in an argon atmosphere and then were examined. No deterioration was noted for tungsten-coated samples having substrates of silicon nitride-bonded silicon carbide, silicon oxide-bonded silicon carbide, Alundum, and Type 430 stainless steel, or for beryllium oxide-coated samples having substrates of Alundum and Type 430 stainless steel. It is planned to obtain test crucibles for evaluation under process conditions.

A thixotropically cast beryllia crucible ($5\frac{1}{2}$ -in. O.D. by 10 in. high) having an "egg-crate"-type bottom, to allow removal of the uranium product in pieces of predetermined size, withstood thermal testing (900°C for 20 hr; 9 cycles between 400 and 800°C) with no apparent adverse effect. Subsequently, a charge of 1.7 kg of uranium was held in the crucible at 1200°C for 2 hr, cooled, and removed as subdivided ingots. In the next test, 3.4 kg of magnesium-zinc-uranium cake having an undetermined amount of graphite on its surface was retorted at 1200°C for 3 hr under

argon to remove the magnesium-zinc alloy. After the run, it was not possible to remove the uranium from the molds in the bottom of the crucible. Such sticking had not previously been experienced in beryllia crucibles following low-temperature retorting operations conducted at less than 1 mm pressure and may have been related to the carbon present or wetting induced by holding magnesium-zinc at high temperature. A slip-cast beryllia crucible having the same dimensions as the other crucible withstood the same thermal testing, but cracked in its first use for dissolution of uranium in magnesium-zinc alloy at 800°C and uranium precipitation.

c. Uranium Oxide Reactions in Molten Halides. Additional information on the thermal decomposition of uranyl chloride in molten chloride media at 650°C has been obtained (see Progress Report for December 1962, ANL-6672, p. 20). The quantity of chlorine evolved by the postulated reaction

was shown to be proportional to the optical absorbance attributed to the uranium(V) species UO_2Cl at 1520 $\text{m}\mu$.

D. FARET

1. Design

An engineering study to determine design feasibility and to estimate cost and schedules for the construction of FARET (Fast Reactor Test Facility) is being performed by Atomics International, Inc., and Shaw, Metz and Associates in conjunction with the Laboratory. The study is approximately 50% complete and the basic concept of the facility and conceptual designs of systems and components have been established.

The FARET Facility is designed to study the neutronics of large dilute fast reactors and to test the performance capabilities of various potential fuels for fast reactors. The reactor is to have the capability of operating at a maximum power of 50,000 kwT with a maximum reactor outlet temperature of 1200°F. Flexibility of operation, accessibility to the reactor for installation of test specimens or special equipment, ability to handle and disassemble spent plutonium fuels having exposures up to 100,000 MD/T, wide range of performance capability of the coolant system, and adequate containment for experiments are the basic requirements. These requirements are reflected in the design of the system herein described.

FARET, as presently conceived, includes the following main features:

- (a) a high temperature, sodium-cooled reactor located within an alpha-gamma type cell;

- (b) a primary coolant system and a decay-heat coolant system located within a shielded vault;
- (c) a secondary coolant system and various auxiliary systems located external to the cell and vault.

All of these systems are housed within the reactor wing of the single-building structure as shown in Figure 3.

Figure 3. Cutaway View of Reactor Wing

One of the principal design features of FARET is the alpha-gamma cell over the reactor vessel. Within the cell are located the reactor control drives, the fuel-handling mechanisms, the fuel-disassembly machine, hot-fuel-storage facilities, and other remote-handling or viewing devices. All operations concerned with fuel handling, loading or unloading the reactor, or installation of special devices into the reactor are conducted within the confines of the cell. In event of an incident, the cell provides shielding for operating personnel and primary containment. The cell has the added feature of assuring a continuing useful life to the facility even if the reactor and primary system were to become seriously contaminated. Operations that would more preferably be done manually would still be possible through the use of manipulators or other remote-handling devices.

The use of a cell over the reactor allows the use of an inert gas atmosphere. As a result, the probability of an air-sodium reaction after an incident is extremely remote. A further advantage of the cell is that the reactor vessel cover may be made the weakest part of the vessel and thereby act as a relief mechanism and serve to attenuate a reactor incident.

The use of a cell offers another attractive feature, namely, it permits use of a conventional type of building. The building becomes a zero-pressure, semicontainment envelope for the controlled release of activity in event of an inadvertent release of activity within the building. The pressure within the building is maintained below ambient. Leakage through the external walls of the building is inward. Exhaust from the building goes to a stack. Should the activity level in the exhaust air become too great, the exhaust is directed through absolute filters and then to the stack.

The top of the reactor vessel is located slightly above the floor in the cell. The vessel cover, associated shielding, and control-rod drives are assembled as a unit on a structure which is supported from and moves on rails. This control-drive complex can be removed from the reactor vessel by disconnecting the drives from the control rods, unbolting the vessel cover, raising the cover and drives above the vessel, and moving the entire assembly to one end of the cell for temporary storage. After removal of the control-drive complex, the top of the reactor fuel assemblies are visible either through cell windows or remote in-cell video units. Normal fuel handling is accomplished by a fuel-handling mechanism which is supported from a crane-bridge-type structure mounted on rails.

The fuel-handling mechanism incorporates a system for cooling the assemblies during transfer from the reactor vessel to the fuel-storage vessel. The fuel-storage vessel is located at the opposite end of the cell from the control-drive storage area. The sodium-filled fuel-storage vessel can accommodate a significant portion of the core assemblies. The sodium in the vessel flows by natural convection to a sodium-to-air heat exchanger for dissipation of decay heat. Disassembly of the fuel assemblies is also provided for in the cell. After fuel disassembly, the various portions of the fuel assemblies are placed in shielded casks for transport to off-site hot cells, reclamation facilities, or disposal facilities, as required.

The cell is provided with personnel-entry locks. During the early life of this facility it is anticipated that considerable personnel access to the cell will be required to install special assemblies, instrumentation, or other devices in the reactor vessel manually. Normally, the reactor vessel will contain sodium at temperatures between 250°F to 350°F. In order to maintain an inert gas atmosphere in the reactor vessel and an oxygen atmosphere in the cell for personnel occupancy, a transparent temporary

cover is used with suitable penetrations for insertion of special equipment into the vessel. The temporary cover is placed over the vessel when the nitrogen atmosphere is in the cell.

The primary sodium system is located entirely within the vault. The vault provides biological shielding and containment in case of a sodium spill. The vault atmosphere is also normally nitrogen. It can be changed to air for the times when personnel entry is required for maintenance or other purposes.

The reactor-coolant systems are designed to be able to perform over a wide range of operating conditions. A brief summary of these conditions are given in Table IX.

Table IX. Primary System Performance Requirements*

<u>Reactor Inlet (°F)</u>	<u>Reactor Outlet (°F)</u>	<u>Bulk Temp Rise (°F)</u>	<u>Power (Mwt)</u>
600 to 960	840 to 1200	240 Max ^b	5 to 50 ^c
300 to 1050	300 to 1050	0 Min	0 Min
1050 Max ^a	1050 to 1200	0 to 150	0 to 31.2 ^d
300 Min	300	0	0

*Raised letters refer to letter notes below.

^aOccasional rise to 1200°F.

^bConsider going as high as 300°F.

^cAssumes 10:1 flow control proportional to power.

^dAssumes maximum flow gives 240°F ΔT at 50 Mwt.

The reactor-coolant system is designed to accommodate a variety of cores. For example, the large cores required for the study of neutronic characteristics may have a pressure drop as little as 1 psi, whereas a small, high-performance core may have a pressure drop as high as 80 psi.

In addition to operating over a wide range of steady operating conditions, slow transient conditions of power and flow are of interest. Control is provided for a power-flow transient such that a constant ΔT across the reactor is maintained. Transients of this type will normally be down-transients.

Removal of decay heat is provided by a separate decay-heat removal loop. A reliable, independent power source is provided to assure removal of decay heat in the event of a power failure. However, as the ultimate heat-removal mechanism, all loops are designed for thermal convection. The decay-heat removal loop is contained within the vault. The secondary portion of the loop dissipates the decay heat to a NaK-to-air heat exchanger.

The secondary sodium system is located exterior to the vault in normally accessible areas of the building. The reactor heat is dissipated to the atmosphere via the primary and secondary sodium systems through a sodium-to-air heat exchanger located outside of and adjacent to the building.

The support wing contains offices, site guard station, locker rooms, miscellaneous utilities, cable-routing room, and the control room. The control room is located in the support wing but adjacent to the reactor wing. The separating wall contains a window to provide the operator visual observation of one face of the cell.

2. Program

A Program Evaluation and Review Technique (PERT) for the planning and controlling of the work in the FARET experimental development program is being established. A basic diagram has been completed (see Figure 4) and will serve as a base to develop a network which will show additional detail of the major activities. The diagram will also provide for the simultaneous development of three core loadings, i.e., (1) a zoned core of 1600 liters; (2) a medium-sized uniform core of 400 liters; and (3) a small high-power-density core of 50 liters.

3. Physics

A medium-sized uniform core of about 400 liters is being considered for the initial experimental program. The program would be directed to measurements on the Doppler coefficient. The proposed core would be fueled with plutonium oxide-uranium oxide mixture having an enrichment of 24.2% ($U^{238}/Pu^{239} \approx 3/1$). A composition of 32/52/16, fuel/sodium/stainless steel structure volume fraction, respectively, was chosen as representative of a typical large fast power reactor. This composition would be typical of a fast power reactor having an enrichment of about 10 to 12%. The core is surrounded by a 30-cm-thick reflector having a composition of 18/82 of sodium/stainless steel volume fraction, respectively. A length-to-diameter ratio of about 0.25 was selected to minimize fuel expansion and isolate the effect of fuel temperature on the Doppler coefficient. Results of computations for the Doppler coefficient on a medium-sized core is shown in Table X. These results indicate that there is a $1/T^\circ K$ behavior pattern. Currently reported values for the Doppler-broadened cross sections have been used in the calculations. It appears that the advantage of the medium-sized uniform core over the zoned loading (driver-buffer system) is of the magnitude of the Doppler coefficient. The advantage is estimated to be about two to three times greater than the value of $\Delta k/\Delta T = 0.73 \times 10^{-6}$ at $750^\circ K$ obtained from calculations for a zoned-core experiment having a U^{238}/Pu^{239} ratio of 7/1. The difference in values is ascribed to volume and statistical weight effects.

Figure 4. FARET Objectives

Table X. Average Doppler Coefficient at Various Temperatures

Temp (°K)	k_{eff}	$\Delta k_{\text{eff}}/\Delta T \times 10^6$		
		(T-300°K)	(T-750°K)	(T-1500°K)
300	1.000001			
750	0.998331	3.71		
1500	0.997075	2.44	1.67	
2500	0.995815	1.90	1.44	1.26

III. GENERAL REACTOR TECHNOLOGY

A. Applied Nuclear Physics

1. Fast-neutron Scattering from W^{184}

This work was undertaken to fulfill a Category-I cross-section request. Some of the preliminary results have been given previously (see Progress Report for December 1962, ANL-6672, p. 24). The measurements are now complete and a formal paper has been prepared.

Neutrons with energies of 0.3 to 1.5 Mev were scattered by W^{184} . Pulsed-beam fast time-of-flight techniques, including a magnetic bunching system, were utilized to resolve the elastically scattered neutrons from those inelastically scattered. The differential elastic cross section was measured at 50-keV intervals with a spread in energy of the incident neutrons of about 20-keV. The differential cross sections for inelastic scattering resulting in the excitation of residual nuclear levels at 111 ± 5 , 365 ± 10 , 690 ± 40 , 900 ± 25 , 1000 ± 30 , and 1120 ± 30 keV were determined. In all instances, the inelastically scattered neutrons were emitted, within experimental error, isotropically. The measured elastic and the inelastic cross sections are shown as a function of energy in Figures 5 and 6.

Figure 5

The differential elastic cross section of W^{184} expressed in the form

$$\frac{d\sigma}{d\Omega} = \frac{\sigma(\text{el})}{4\pi} \left[1 + \sum_{i=1}^5 W_i P_i \right]$$

The solid points refer to measurements of the elastic component only. Open data points represent measurements which included its fast inelastically scattered neutron group with the elastic component.

2. Fast-neutron Spectroscopy

The development of lithium-sensitized solid-state neutron spectrometers having improved energy resolution was last reported in ANL-6573 (Monthly Progress Report for May, 1962, p. 27). The improvement in resolution is achieved primarily by providing a moderate degree of collimation for the reaction products from the lithium.

Figure 6. The Inelastic Cross Section of W^{184} .

spectrometer was accepted in order to optimize the energy resolution. The pulse distribution observed with this instrument in a thermal-neutron flux showed a sum-pulse line of full width at half maximum of only 85 keV, corresponding to an energy resolution of 1.8%.

A more flexible instrument, for use with the ZPR-VI, has also been built. It employs a thin plastic film to support the sensitive lithium layer, which is evenly spaced between the two diodes. Pulse distributions in a helium atmosphere and in vacuo were measured in order to explore the possibility of building a spectrometer to work in helium and eliminate the relatively bulky pump connections.

A set of data has been taken with the more sensitive instruments in the reflector of the AFSR. The pulses derived from fast neutrons appear to be superimposed upon a background of unknown origin. With the hope of

In recent months, a number of these fast-neutron spectrometers, employing the $Li^6(n, \alpha)H^3$ reaction and two solid-state detectors in coincidence, have been built, tested, and sent to the Idaho site for making measurements of fast-neutron spectra. Two instruments were designed for use with ZPR-III and, hence, required good neutron efficiency. The width of the pulse distribution at half maximum for these detectors having a relatively thick lithium layer was approximately 4%. A third spectrometer was designed to make measurements in the glory hole of the AFSR, and therefore was limited in diameter to less than 1.2 cm. A lowered efficiency in the

eliminating such background through a coincidence arrangement, a spectrometer model permitting two separate outputs has been designed. The mechanical parts have been made but completion of the instrument is delayed pending receipt of a set of suitable solid-state detectors.

3. High-conversion Critical Experiment (ZPR-VII)

In ANL-6683 (Monthly Progress Report for January 1963, p. 23), a series of measurements made on less undermoderated assemblies was described. These measurements were confined to a determination of critical mass and reflector savings, and are intended to provide an overlap between the results of the highly undermoderated assemblies obtained in the Hi-C program and experimental data obtained elsewhere on assemblies having a higher degree of moderation. Buckling measurements were made with an assembly having one of every eight fuel pins removed and were followed by measurements made with one in which every fourth fuel pin was removed. The uniform lattice in this 1.24-cm square pitch pattern has now been restored for a repetition of some of the microscopic lattice-parameter measurements suspected of lacking the desired accuracy.

Two recent developments in the Hi-C program are worthy of note. In ANL-6683 (Monthly Progress Report for January 1963, p. 23), it was noted that there was a rather thorough modification in the data-reduction program used in the analysis of foil-activation data. As a result of these modifications, the reproducibility of the buckling measurements has been greatly improved, and this is, in part, attributable to the correction of a procedural error in assigning statistical weights to the data used in the COFIT and JOFIT programs.

The second item of interest is the successful demonstration of the strong absorption in the energy region of the U^{238} resonance for assemblies having high fuel-to-moderator ratios. These results have been obtained from the activation data resulting from measurements with indium, gold, manganese, and copper. Indium and gold have resonances in the 1- to 5-ev region, whereas manganese and copper have resonances in the 300- and 600-ev region. Were it not for significant absorption in the interlying energy region, the product of the energy and the flux at that energy would be expected to be approximately a constant. Data obtained for the tightly packed triangular Hi-C lattice (1.13-cm triangular pitch) demonstrated that this product was approximately twice as large in the 300- to 600-ev range. Similar results have been obtained with the BORAX-V superheater criticals studied in the ZPR-VII system. Measurements in the tight lattice and peripheral boiling zone of the BORAX-V system are consistent with the variation in the fuel-to-moderator ratio.

4. Theoretical Physics

a. Numerical Methods Analysis. Considerable savings in computing time may be effected by the judicious selection of approximations to evaluate frequently occurring functions. A general routine has been written for the LGP-30 computer to find the best approximation (in the sense of minimizing the maximum error over a specified range) among the class of functions of a particular form which depend upon a given number of parameters. The form of the approximating function may be any one which can be generated by an interpolation routine.

Approximation of the natural logarithm by polynomials was the first example explored. To make the error small enough for round-off to be negligible, the (impractically large) range $1 \leq x \leq 20$ was chosen. It was found possible to fit $\ln x$ by a polynomial in x over this range with maximum errors of 0.224, 0.110, 0.057, and 0.022, for third, fourth, fifth, and sixth-degree polynomials, respectively. It was then decided to investigate fitting the function $\ln(1 + ax + x^2)$ over a range of x such that $1 \leq 1 + ax + x^2 \leq 20$. This is equivalent to fitting $\ln x$ over the range $0 \leq x \leq 20$ by a cubic polynomial in $\sqrt{[(a^2/4) - 1 + x]} - a/2$. For $a = 0.8892$, the maximum error was less than 0.019, appreciably better than that of the polynomial of twice the degree in the original variable. This example illustrates the importance of finding the optimum independent variable to use in an approximation problem.

B. Reactor Fuel Development

1. Ceramic Fuels

a. UO₂. Two types of fuel have been suggested for the measurement of the Doppler effect in FARET. The first type consists of dense UO₂ grains bonded with a small amount of glass. Pellets, 2.54 cm in diameter by 3.18 cm in length, were cold pressed with 1 w/o sodium silicate as a binder. These pellets, after sintering at 1200°C in hydrogen, were found to have a density of 70% of the theoretical density of UO₂. Eleven of these pellets were pressed into a metal tube fitted with a resistance heater for initial out-of-pile experiments.

The second type of fuel consists of extruded UO₂ rods having three longitudinal holes spaced at 120° around a central hole. The outer surface of the rod has small projections which serve to center the fuel in its jacket and to allow space for gas passage through the element. An extrusion die and a core bridge have been made. Initial extrusions showed a tendency to crack on drying; however, by reducing the water and binder content slightly, good UO₂ extrusions could be fabricated.

b. Plutonium and Plutonium-Uranium Carbides. Pellets were made from powder that was prepared by crushing arc melted buttons of PuC containing 46.5 a/o carbon. The pellets were chemically analyzed for carbon, oxygen, and nitrogen contents and spectrographically analyzed for other elements, particularly iron.

The oxygen analysis showed that there was an increase in oxygen from 0.0050 w/o in the arc-melted button to 0.2 to 0.4 w/o in the pellets. There was no significant pickup of nitrogen resulting from handling the powders in the nitrogen atmosphere of the gloveboxes, as shown by the low nitrogen content in the pellets of about 0.0150 w/o. The concentrations of carbon and other elements (determined by spectrographic analysis) were about the same as found in the arc melted button before crushing. Powder has been stored in sealed containers surrounded by a desiccant for periods of a month without an apparent increase in oxygen content as would be evident by a green color in the powder.

Six pellets were made from arc-melted material containing 50.0 a/o C to see if this powder, which contained about 25% of the Pu_2C_3 phase, would sinter in the same manner as the pure PuC powder containing 46.5 a/o carbon. The pellets were mixed with various percentages of Carbowax ranging from 0.5 to 4 w/o, pressed at 80,000 psi and sintered at 1400°C for two hours. As found with the 46 a/o C material, there was a gradual decrease in density from about 93% of theoretical with increasing Carbowax content.

A series of three plutonium carburization melts were made to determine whether the recently installed equipment for temperature control would produce carbide of more uniform quality. The temperature-control cycle at 1800°C was reduced to approximately $\pm 10^\circ\text{C}$ by means of the radiation pyrometer and automatic control, as compared with $\pm 40^\circ\text{C}$ control attained with the optical pyrometer and manual control method. Samples from these melts have been submitted for analysis.

A uranium-plutonium carbide melt was prepared by reacting 80 w/o U-20 w/o Pu alloy with carbon. The melt temperature was carried to approximately 2100°C. Samples from this material are presently under analysis.

The furnace used in this work has been fired to over 2000°C for a total of ten times and to 1800°C approximately 30 times. The two inner tantalum heat shields have deteriorated and failed during the last melt. The molybdenum heat shields, which operate at a somewhat lower temperature than the tantalum shields, have not shown deterioration. Some of the internal stainless steel supporting members were also melted during the last heat. These stainless steel members are being replaced with TZM alloy.

c. Uranium-Thorium Sulfide. An attempt to improve the sintering characteristics of US by ball milling to finer particle sizes in an argon-filled mill in air was unsuccessful because of severe oxidation of the powders. The work did, however, establish that US does not become pyrophoric until an average particle size of 0.8μ is reached. The milling study will be rerun inside an argon-atmosphere box.

Two different methods of eliminating the undesirable UOS phase from US were tried. In the first, US pellets which had previously been sintered to 91% theoretical density in argon and contained about 3 w/o UOS, were refired in vacuum at 1825°C for 6 hr. Polished sections showed no sign of either UOS or UO_2 secondary phases, and would seem to indicate that UOS vaporizes congruently at that temperature. The second method utilized additions of free carbon to the US powder before compacting on the theory that the carbon would reduce any UOS present to $\text{US} + \text{CO}$, and any excess carbon would go into solid solution. Compacts of US containing 1, 2, 3, 4, 5, 7, and 9 m/o C fired at 1825°C for 3 hr in argon showed a slight decrease in UOS content to about 2 m/o C, but a progressive increase in the amount of secondary grain boundary phase for higher carbon additions. At high magnification, an increasing amount of a darker gray phase was found to coexist with the UOS at the grain boundaries. Since the lattice parameter of the US showed only a very slight decrease, it is probable that this gray phase, as yet unidentified, contains the excess carbon.

An investigation of the US-UO_2 system was initiated in order to better understand the overall effects of oxygen contamination in US. Compacted mixtures of US with 5, 10, 20, 40, 60, and 80 m/o UO_2 were fired in vacuum at 1825°C for 3 hr. The bodies were evaluated by X-ray powder patterns and polished sections. The UO_2 was found to occur as discrete granular areas which became more rounded with increased UO_2 content. No oxygen from the UO_2 went into solution in the anion positions of the US structure, since the US lattice parameter remained essentially constant. A eutectic seemed to occur between US and UO_2 at about 90 m/o UO_2 . This helps to explain why vacuum-fired US containing UO_2 gets little aid in densification from liquid-phase sintering. It is planned to examine the US-UOS system similarly.

Sintering US specimens in an argon atmosphere was found to cause a decrease in the US lattice parameter because N^{-2} ions went into solid solution in its cubic structure. The extent of this nitrogen solubility was investigated by firing compacted mixtures of US with 2.5, 5, 10, 20, 40, 60, and 80 m/o UN at 1825°C in vacuum for 3 hr. X-ray diffraction patterns showed a progressive decrease in lattice parameter from 0 to 10 m/o UN, but no further decrease for larger UN contents. The UN lattice parameter towards the high UN contents showed no perceptible change, indicating

negligible solubility of US in UN. Although equilibrium was probably not quite attained in this firing, it is unlikely that more than 10-12 m/o UN is soluble at this temperature. Even this amount of solubility is somewhat surprising since the ionic radius of N^{-2} (1.569 Å) is some 17.5% smaller than that of S^{-2} (1.903 Å)

The solubility of ZrN and HfN for 5 and 10 m/o additions was also determined in the same firing at 1825°C. These additions offer good potential for raising the melting point of US since both melt at greater than 3000°C. X-ray patterns indicated the solid solubility of from 5-10 m/o ZrN and 2-5 m/o HfN in US, although polished sections did not show the presence of the nitride phases

d. Uranium, Thorium, Plutonium Phosphides. The objective of this study is to characterize the compounds in the different phosphide systems and evaluate their potential as reactor fuel materials. Compounds are currently being synthesized by reaction of the elements, although other methods will be explored.

Experiments have continued involving the heating of oxygen-contaminated UP in vacuum at elevated temperatures. A large sample in a tungsten crucible showed a 2.8% weight loss in an hour at 2200°C. Part of this loss was of UO_2 , since X-ray analysis of the condensate showed it to consist of 35% of UO_2 , whereas the original material contained about 10% UO_2 . Heating of the same material at 2400°C resulted in a loss of weight at the rate of about 21% per hour. Metallographic analysis of the latter material still showed the presence of two metallic-appearing phases, in addition to discrete particles of UO_2 . One of these phases has been identified as uranium metal and the second is presumed to be a substoichiometric UP phase. Based on these preliminary results, it appears that UP exhibits a moderate dissociation pressure at elevated temperatures, with phosphorus being driven off preferentially. Upon cooling, the excess uranium is expelled from the sodium chloride-type structure in the form of free uranium and/or uranium-rich UP. Whether these phases can be retained in the structure by rapid quenching will be examined in future experiments.

X-ray analysis of compositions across the UP- UO_2 system shows that the UO_2 structure is unaffected by the presence of UP during heating at 2000°C. The lattice constant of UO_2 remains unchanged at 5.473 ± 0.005 Å across the entire compositional range from pure UO_2 to 90% UP-10% UO_2 . UP shows virtually no change in its lattice constant from compositions containing 90% UP to those containing 10% of the compound. The structure of pure UP is not yet known because it has not been possible to prepare a sample which does not contain UO_2 .

Metallographic examination of UP powders subjected to a flowing stream of oxygen in the DTA apparatus revealed that the compound is not completely oxidized until above 700°C. A coating, presumed to consist of UO_2 interspersed in a vitreous uranium phosphate glass, surrounds the UP grains and gets thicker with increasing temperature (and time). Only small amounts of UP remain after heating to 700°C. The oxidized grains show signs of cracking after being heated to 800°C, probably a result of oxidation of the interspersed UO_2 to U_3O_8 (which occurs at about 750°C). The grains develop strains and shatter during polishing after being heated above 900°C, owing to devitrification of the glassy phase. Devitrification is corroborated by X-ray analysis, which shows the appearance of a new crystalline phase above 800°C.

Considerable effort has been given to finding a satisfactory organic binder for use in cold pressing of UP samples. Over ten different binder and binder-lubricant combinations have been studied thus far. Nearly all of them produce superficial cracking, which does not appear to be related to pressing pressure. The best binders to date are sodium alginate and acryloid-stearic acid. The effect of aging on crack formation is currently being studied, as it appears to be significant.

e. Plutonium Sulfide and Phosphide. During the past month several attempts were made to make PuP by different techniques. Plutonium metal was hydrided at about 150°C and then dehydrided at 350°C in a vacuum of about 10μ to get plutonium metal powder. This powder was then mixed with red phosphorus powder and heated; however, no reaction took place. Either the plutonium was not completely dehydrided or plutonium does not react with phosphorus at the relatively low temperature used. Therefore, plutonium metal pieces and phosphorus powder were placed in a closed container and rapidly heated to 1000°C in an inert atmosphere of argon gas. Most of the phosphorus escaped from the container at about 700°C, and only a surface reaction was observed on the metal pieces. A bomb reaction will be tried soon.

Plutonium metal was successfully combined with phosphorus in the arc furnace. However, this combination was achieved only with great difficulty because of the vaporization of the phosphorus. Unlike the spontaneous reaction of plutonium metal with sulfur, the metal-phosphorus reaction does not appear to be vigorous and self-sustaining.

f. Rare Earth Oxides as Burnable Poisons. The objective of this study is to stabilize rare earth oxides in perovskite-like compounds (ABO_3) by combining them with other oxides. The two rare earth oxides of interest are Eu_2O_3 and Dy_2O_3 , which are being combined with suitable oxides of cobalt, iron, and nickel.

High-purity Eu_2O_3 and Dy_2O_3 powders were calcined at 1000°C and each mixed in proper proportions with reagent-grade powders of Co_3O_4 , Fe_2O_3 , and NiO , which had been heated to 500°C . The powders were blended in a ball mill and pressed into small pellets using an organic binder.

Samples were fired in air at 1400°C for 2 hr with a slow heating schedule. With the exception of $\text{Co}_3\text{O}_4 \cdot \text{Eu}_2\text{O}_3$, which melted and reacted with the alumina crucible, the pellets retained their original shapes. X-ray analysis showed complete reaction of Fe_2O_3 with both Eu_2O_3 and Dy_2O_3 to form FeEuO_3 and FeDyO_3 . FeEuO_3 has been described in the literature, but FeDyO_3 is a new compound. CoEuO_3 appeared to have formed, but it apparently melts below 1400°C . No sign of reaction was observed between NiO and either Eu_2O_3 or Dy_2O_3 or between Co_3O_4 and Dy_2O_3 .

Additional blended samples of Co_3O_4 and NiO and the two rare earth oxides were fired for $3\frac{1}{2}$ hr at 1200°C . None of the pellets melted at this temperature. X-ray analysis revealed almost complete reaction between Co_3O_4 and Eu_2O_3 to form CoEuO_3 . This compound is isomorphous with FeEuO_3 and also has never been reported before. Incipient reaction occurred between Co_3O_4 and Dy_2O_3 , but none was observed between NiO and either Eu_2O_3 or Dy_2O_3 .

DTA studies revealed that the reaction between Fe_2O_3 and Eu_2O_3 is exothermic and occurs at about 1200°C . The reaction between cobalt oxide and Eu_2O_3 is also exothermic and takes place at about 1060°C . Actually, Co_3O_4 dissociates to CoO at 950°C and it is the latter compound that reacts with Eu_2O_3 .

The cubic to monoclinic transformation of Eu_2O_3 at about 1050°C may account for its greater reactivity compared with Dy_2O_3 . The latter compound does not undergo a similar transformation, at least under 1300°C . The greater stability and refractoriness of NiO compared with Co_3O_4 and Fe_2O_3 may account for its lesser reactivity.

2. Radiator Fuel Elements

A welding test specimen was made up to demonstrate the feasibility of fabricating miniature heat-exchanger shells. It consists of a hexagonal tube (made from EBR-II hexagonal tube stock) containing fifty-eight 0.156-in.-ID by 0.009-mil-wall-thickness tubes and three filler tubes. The tubes were TIG-welded to two 0.040-in.-thick tube sheets. The assembly was held on two rotary feed tables mounted "piggy back" one above the other. The lower table was driven by a variable-speed gear motor. The upper table was fitted with a cross-feed device which allowed accurate positioning of the welds below a welding torch as the assembly was rotated. The welding current was

programmed to the rotary drive to control lapping and cratering. This first test sample, made from available tubing stock, was successful and hardware was ordered to try a 91-tube test assembly.

3. Corrosion Studies

a. Structural Materials in Superheated Steam. In a five-week dynamic test Type 304 stainless steel corroded faster than in a static exposure (see Monthly Report for January 1963, ANL-6683, p. 32). The experiments were performed in oxygenated (30 ppm) steam at 650°C, 600 psig and at velocities of 100, 200 and 300 ft/sec. The defilmed metal-loss rate decreased after the first week and was approximately linear at about $3\frac{1}{2}$ mils/yr for the next four weeks in the dynamic test. This may be compared with a rate of about 1 mil/yr measured in the previous static test.

Type 406 stainless steel showed no effect of velocity, and its corrosion rate after the first week was too low to be determined by the techniques used. About 0.0001 in. (average) of metal was corroded on all the specimens, whether for one week or five.

Currently, Types 304 and 406 stainless steel are being tested dynamically in deoxygenated steam at the same conditions of temperature, pressure, and velocity.

Rockwell hardness measurements were made of samples of Zr-3 w/o Ni-0.5 w/o Fe and Zr-1 w/o Cu-1 w/o Fe steam-resistant alloys after corrosion in deoxygenated steam at 650°C and 600 psi for 88 days in order to observe possible embrittlement of the metal due to absorption of corrosion-product hydrogen or oxygen. The results were compared with similar measurements made before corrosion test. In the nickel alloy little or no change of hardness occurred; the material remained relatively soft at an RC value of about 13. In the copper alloy, which is hardenable by quenching (to RC 33), a quenched sample was softened during corrosion exposure to RC 8, a value close to that of the original unquenched material.

Samples of the above materials and a Zr-1.1 w/o Cu-0.7 w/o Fe alloy were exposed to steam flowing at 100, 200, and 300 ft/sec, conditions otherwise as above. During the first week of exposure, the corrosion behavior was similar to that in static tests; the dynamic test is continuing.

It is apparent that Zr-4 a/o Cu and Zr-4 a/o Ni binaries are not critically sensitive to startup procedure in 540°C steam tests. However, pretreatment at 700°C in oxygen for 16 hr was deleterious to the steam resistance of Zr-Ni. An unexplained steam test result was obtained for Zr-Cu pretreated in oxygen. A continuous area covering about $\frac{1}{2}$ of the sample was poor relative to untreated material, while the other half was superior in appearance. This condition has not changed appreciably during exposure for the past month.

b. Miscellaneous Materials in Water and Steam. One sample of TD nickel developed a number of surface blisters during corrosion test (see Progress Report for January 1963, ANL-6683, p. 33). To test whether or not the blistering was associated with local concentrations of thoria, an X-ray film was placed in contact with the TD material, and a self-exposed image was obtained. No pattern comparable to the blister pattern was detected, indicating that the blistering was probably not due to segregated thoria.

4. Fuel-jacket Development for High-temperature Applications

Two Ta alloys, Ta-10 w/o W and Ta-8 w/o W-2 w/o Hf, were received in $\frac{3}{4}$ -in.-diameter bars for the purposes outlined in the Progress Report for January 1963, ANL-6683, p. 33. Portions of both alloys have been cold rolled and drilled into tubes of the following approximate dimensions: 0.440 in. in OD x 0.312 in. in ID x 5 in. long. The tubes are to be annealed and drawn down into 0.156-in.-OD x 0.015-in.-wall tubing.

5. Nondestructive Testing

a. Ultrasonic Imaging. Life-test studies on a sealed-off ultrasonic pickup tube having a 2-in.-diameter, 2-megacycle quartz target are still in progress. In addition to cathode-emission studies, the tube is now being subjected to ultrasonic radiation in order to determine what effect this may have on the target seal. A second sealed-off tube has been obtained, and life-test studies have begun.

Equipment for setting up an ultrasonic imaging system has been received and is now being assembled.

b. Correlation of Heat Transfer Properties and Bond Quality. Work described in the Progress Report for October 1962 (ANL-6635, p. 39) has been continued. Redesign of the test blocks with a brazed bond which had been prepared for use in these studies has been planned. This present set of specimens was made without control of factors which can affect the strength of the bonds, e.g., surface preparation, pressure used when making the bond, and thickness of the intermediate layer. Control of these factors is required if the strength properties of the specimens are to be correlated with the presence of defects in the bond. The thickness of the elements to be bonded must also be considerably reduced to prevent too great a conduction and diffusion of heat from giving a temperature distribution on the surface different from that occurring directly over the bond layer.

A review of the various methods for heating specimens of this type has shown that either the "hot plate" apparatus designated by ASTM: C411-58T or the "guarded hot plate" apparatus designated by ASTM: C177-45 used for thermal conductivity measurements can be adopted for these heat transfer studies.

c. Development of Improved Transducers. Work continued this month on the fabrication of operational transducers. The major effort was concentrated on improvement of backing materials, although considerable time was spent in the preparation of 5-Mc crystals from 1-Mc blanks. Only 5-Mc lead zirconate-titanate crystals were utilized in studies this month.

As a further comparison of the relative merits of tungsten-loaded and fiberglass-loaded epoxies as backing materials, two transducers were fabricated as identically as possible, the type of backing material being the only difference. Both backing materials extended around the edges as well as behind the crystal. These transducers produced pulses of the same approximate length.

One transducer faced with a silver-loaded epoxy successfully passed a one-month immersion test in water. The transducer still functioned normally when applied to a test specimen.

A gaging system has been built to check on the flatness of the crystal when mounted in the transducer housing. It will also be used to check incoming crystal blanks.

d. Radiographic Techniques. Work on alpha spectrometry as a means of isotopic analysis has continued. An electroplating cell with a variable-speed rotating cathode has been constructed for electroplating uranium on platinum disks. Several samples of good quality were produced by this method. The sample was deposited on both sides of the platinum disk with approximately twice as much being deposited on the side toward the anode. By using two detectors in parallel, both sides were counted simultaneously, but the resultant spectra showed very poor resolution. By coating one side of the platinum disk with a nonconducting material it should be possible to quantitatively deposit uranium on one side only.

e. Application of Infrared Radiation. Work was initiated on an investigation of the possible use of infrared radiation as a nondestructive testing tool. In the summer of 1960, some preliminary investigations were conducted with indium antimonide photoelectric-magnetic detectors. At that time no attempt was made to develop a suitable optical system for use with the detector and source. Therefore, the first objective of this investigation is to assemble an optical system.

A literature search has also been started to determine the extent to which infrared technology has been applied to nondestructive testing.

6. Extrusion Development

Work continued on developing the DPM extrusion technique (see Progress Report for November 1962, ANL-6658, p. 48). The high degree of wall rippling evidenced in a series of 15 extrusions has led to a modification of the technique. The modification consists of placing a metal core in the extrusion billet with the annulus between the wall and core being filled with a particulate material. The addition of the metal core tremendously improves wall uniformity and concentricity of the extruded products, making it suitable for secondary fabrication. Additional work-ing can be done with an empty tube (metal core and powder removed) or with the composite tube with the metal core serving as a ductile core to be removed at or near the final fabrication operation. Core removal from extruded tube blanks and secondary drawing of the composite tube have been easily accomplished.

Extrusions of modified DPM billets have included copper jackets with copper cores and Zircaloy-2 jackets with copper, Zircaloy-2, and Armco iron cores. While the wall thickens slightly with respect to diameter, owing to compression of the powder, wall uniformity in both the longitudinal and transverse direction is excellent. Some degree of ovality occurred in the Zircaloy-2 extrusions but was not serious. Further fabrication of these extrusions is planned as well as additional modified DPM extrusions using stainless steel as the billet material.

C. Heat Engineering

1. Heat Transfer in Double-pipe Heat Exchangers

The analytical study of heat exchange between fluids of low Prandtl number flowing through adjacent parallel plane channels (see Progress Report for November 1962, ANL-6658, p. 50; Progress Report for October 1962, ANL-6635, p. 41) is being continued.

Analysis of the case of cocurrent plug flow has been completed and includes solutions for flow over the entire duct length. A computer program is available to provide numerical values for the various quantities necessary in presentation of the solution. In addition, the analysis has been generalized to provide numerical results for more realistic (cocurrent laminar and turbulent flow) cases. It is planned to investigate the use of an approximation method to solve the simpler cases of cocurrent plug flow.

In preparing to evaluate the turbulent flow case, methods of computing eddy diffusivities were reviewed. Figure 7 shows a comparison of the two most frequently used methods for computing the eddy diffusivity of

momentum with correlations developed from measured values for mercury and air. The agreement between the two correlations of data is especially interesting because of the large differences between the physical properties of the fluids, viz., a heavy liquid metal and a gas.

LEGEND FOR FIGURE

- A SMOOTHED DATA OF PAGE, F. ET AL, IND. ENG. CHEM. 44, 424 (1952): AIR IN PARALLEL PLANE CHANNEL
- B DATA CORRELATION OF ISAKOFF, S.E. & T.B. DREW, AECU-1199, (1951): MERCURY IN ROUND TUBE, CONVERTED TO VALUES APPROPRIATE TO PARALLEL PLANE CHANNEL
- 1 CALCULATED VALUES USING VON KÁRMÁN'S RELATIONS
- 2 CALCULATED VALUES USING DEISSLER'S RELATIONS
- ϵ_m = EDDY DIFFUSIVITY OF MOMENTUM
- ν = KINEMATIC VISCOSITY OF FLUID

Figure 7. Eddy Diffusivity Distributions in Parallel Plane Channel at a Reynold's Number of 52,500 Comparisons of Data with Calculations.

Effort on the solution of the case of countercurrent plug flow is being continued. Iteration-approximation techniques have been outlined, based on use of analytical solutions of heat transfer in single channels with arbitrary wall temperature and/or heat flux distributions.

D. Chemical Separations

1. Chemical-Metallurgical Process Studies

a. Chemistry of Liquid Metals. The skull material remaining after the melt-refining step will contain fission product zirconium and fragments of the zirconia melt-refining crucible. The removal of some

of this zirconium occurs during the skull-reclamation process step in which a uranium-zinc intermetallic compound is precipitated from a 5 w/o magnesium-zinc solution by cooling the solution from 800 to 500°C. The extent of removal of zirconium depends on the solubility of zirconium in the 5 w/o magnesium-zinc solution at 500°C. Zirconium in excess of its solubility value under these conditions will precipitate out of the solution and will accompany the uranium-zinc intermetallic compound.

To establish the amount of zirconium that can be removed in this process step, the solubility of zirconium in 5 w/o magnesium-zinc solution was measured. The solubilities ranged from 0.06 w/o zirconium at 480°C to 1.1 w/o at 600°C. The solubility at 500°C was estimated by interpolation to be 0.11 w/o. This value allows the calculation of the percentage of zirconia crucible fragments that can be tolerated (about 0.8 w/o) in the skull material, assuming that the equilibrium concentration of zirconium in recycled EBR-II fuel will be maintained at about 3 w/o and assuming also that the zirconium is removed only in the uranium-zinc intermetallic precipitation step at 500°C.

The thermodynamics of the plutonium-zinc system are being studied by galvanic cell method. Emf data for the cell Pu/PuCl₃, LiCl-KCl (eutectic)/Pu-Zn (2-phase alloy) have been obtained over the temperature range from 421 to 615°C. Two recently constructed cells were used in these studies. The second cell differed from the first in two respects: the anode consisted of a plutonium rod welded to a tungsten lead wire instead of plutonium contained in a thoria crucible, and the PuCl₃ content of the lithium chloride-potassium chloride eutectic was 0.3 w/o instead of 3 w/o. These changes had no effect on the emf of the cells, and the data obtained with the two cells were found to be in excellent agreement (see Figure 8). The following equation was derived for the range from 420 to 670°C by combining the data obtained with the two cells:

Figure 8. Emf of Cell: Pu/PuCl₃, LiCl-KCl (eutectic)/Pu-Zn (2-phase alloy) vs. Temperature.

$$E \text{ (v)} = 0.95906 - 0.36640 \times 10^{-3} T - 0.16845 \times 10^{-6} T^2$$

The standard deviation was found to be 0.56 mv. At 450°C, the following preliminary values pertaining to Pu₂Zn₁₇ have been calculated: the free

energy of formation (ΔG_f°) = -42.0 kcal/g-atom Pu; the entropy (ΔS_f°) -42.2 cal/(deg) (g-atom Pu); and the heat of formation (ΔH_f°) = -72.5 kcal/g-atom Pu.

A preliminary measurement of the magnetic susceptibility of uranium monosulfide was made at room temperature. The tentative value for the susceptibility was calculated to be 22.3×10^{-6} emu/gram or 5.9×10^{-3} emu/mole.

b. Preparation of Uranium Carbide. The reaction of methane with uranium-zinc intermetallic compounds is being studied as a possible method for the synthesis of uranium carbide. The reaction rate has been found to increase with temperature, the partial pressure of methane, and the gas flow rate. The reaction rate remained approximately constant until the number of moles of methane reacted per gram atom of uranium present reached a value of about one or higher; then the rate decreased markedly. The reaction product was a loose, fine powder which was not homogeneous. Homogenization at 1500 to 1800°C is required to obtain a uniform product. For comparison, uranium metal which had been powdered by hydriding and dehydriding was reacted with methane. The reaction rate for the uranium metal-methane reaction was significantly lower than the rate of reaction of methane with the uranium-zinc intermetallic compounds under comparable conditions.

2. Fluidization and Fluoride Volatility Separations

a. Fluoride Separations. The effect of radiation on the decomposition of plutonium hexafluoride is being studied to provide information which is of fundamental interest in the development of processes for the separation of plutonium hexafluoride from uranium hexafluoride. Additional data have been obtained on the gamma radiation decomposition of plutonium hexafluoride to plutonium tetrafluoride and fluorine in the presence of helium and krypton (see Progress Report for January 1963, ANL-6683, p. 40). Experiments on gamma irradiation of plutonium hexafluoride samples (80 mm Hg pressure) containing helium at one and two atmospheres have resulted in G values* of 3.4 and 1.8, respectively. The total absorbed energy dose for each run was about 40×10^{20} ev. In analogous experiments on the gamma irradiation of plutonium hexafluoride in the presence of krypton, a similar decrease in the G value was obtained at the higher pressure of krypton. However, a high G value of 8.9 was obtained in an experiment in which the plutonium hexafluoride sample was irradiated in the presence of one atmosphere of helium but in which the total absorbed energy dose (9.1×10^{20} ev) was considerably lower. Further experiments are planned to study these effects.

Additional data have been obtained on the gamma irradiation of plutonium hexafluoride samples (80 mm Hg pressure) in the presence of krypton. In two experiments made with krypton at one atmosphere and with high total absorbed energies (80×10^{20} and 106×10^{20} ev), the G values were 1.4 and 1.0, respectively. These results confirm the marked decrease in G value (0.6) which was noted in a similar experiment with fluorine in which the total absorbed energy dose was 69×10^{20} ev. In one experiment made with krypton at one-quarter atmosphere, the G value was 3.7, which is not markedly different from the G value obtained in an experiment made with krypton at one-half atmosphere. In one experiment made with krypton at 20 mm Hg pressure and a total absorbed energy dose of 16×10^{20} ev, the G value was 6.4. This G value of 6.4 is close to the average G value of 7.4 obtained for the gamma radiation decomposition of plutonium hexafluoride when irradiated alone. The data obtained in these studies are summarized in Table XI.

Table XI. Decomposition of Plutonium Hexafluoride by Gamma Radiation in the Presence of Various Gases

Pressure of Plutonium Hexafluoride: ~80 mm at 25°C

Gas	Pressure (atm)	Total Absorbed Energy (ev $\times 10^{-20}$)	G Value*	Gas	Pressure (atm)	Total Absorbed Energy (ev $\times 10^{-20}$)	G Value*
Helium	1	9.1	8.9	Krypton	0.25	25	3.7
Helium	1	40	3.4	Krypton	0.5	34	4.3
Helium	2	40	1.8	Krypton	0.03	16	6.4
Krypton	1	80	1.4	Fluorine	1	69	0.6
Krypton	1	106	1.0				

*Moles decomposed per 100 ev of radiation.

b. Direct Fluorination of Uranium Dioxide Fuel. The development of a two-zone oxidation-fluorination scheme for the conversion of uranium dioxide pellets into uranium hexafluoride in a single reactor was continued in engineering-scale studies (see Progress Report for January 1963, ANL-6683, p. 41). In this scheme, U_3O_8 fines are produced in the lower reaction zone by contacting the uranium dioxide pellets in this zone with an oxygen-nitrogen mixture. The U_3O_8 fines are elutriated from the pellet bed and fluorinated with fluorine in the upper reaction zone. Alumina is present in both zones as a fluidizing medium. The transport of U_3O_8 fines from the uranium dioxide pellet bed to the fluorination zone is facilitated by the presence of the fluidized alumina.

Previously reported (ANL-6683) two-zone oxidation-fluorination runs were made to investigate the effect of temperature gradients and gas pulsing in the pellet bed on the production and transport of U_3O_8 fines. These techniques were intended to ensure rapid transport of fines from the lower oxidation zone to the upper fluorination zone. The results of these runs have shown the following:

- (1) The production of large amounts of U_3O_8 fines and the transport of these fines to the fluorination zone require dilution of fluorine with inert gas to prevent plugging of fluorine inlet lines to the reactor by intermediate uranium fluoride compounds.
- (2) Gas pulsing provides a significant improvement in U_3O_8 fines transport from the pellet-bed oxidation zone to the fluorination zone.
- (3) High concentrations of oxygen (about 20 percent) and temperature gradients in the pellet-bed oxidation zone are not required for the high-rate production of uranium hexafluoride.

On the basis of these results, two runs were made to study further the effects of gas pulsing, of maintaining the same temperature in the oxidation and fluorination zones, and of low concentrations of oxygen in the oxidation zone. In each of these runs a 12-in.-deep static pellet bed (8.8 kg uranium dioxide) and a 20-in.-deep fluidized alumina zone (7.1 kg) above the pellet charge were used. The pulse duration was 0.04 sec with a pulse frequency of 2 pulses/min. The temperature in the two reaction zones in each of the runs was about 500°C. In the oxidation zone, the compositions of the fluidizing gases in the two runs were 4.5 or 6.0 percent oxygen in nitrogen. The uranium dioxide charges in the two runs were completely fluorinated in total processing times of 15 and 12.5 hr, respectively, and with overall fluorine efficiencies of about 70 percent for both runs. The total processing times included a period of alumina-bed cleanup

with fluorine. The shortest processing time (12.5 hr) achieved thus far was established in the run in which 6.0 percent oxygen was used. Sustained periods of high rates of hexafluoride production were attained, and a maximum production rate of 107 lb UF_6 /(hr)(sq ft of reactor cross section) was demonstrated.

c. Fluorination of Uranium Oxide (U_3O_8). Uranium oxide (U_3O_8) is formed as the major product in the oxidative decladding of uranium dioxide pellet fuel elements. The kinetics of the fluorination reaction of uranium oxide are therefore of interest in the fluid-bed fluorination of this oxide (see Progress Report for September 1962, ANL-6619, p. 47). Data for this fluorination reaction have been measured for the temperature range from 300 to 400°C by means of a thermobalance. Samples of uranium oxide from two sources were fluorinated, a standard sample (-100 +325 mesh) of certified purity from an AEC laboratory and a sample (-140 +200 mesh) from a commercial source. A diminishing-sphere model was used in the treatment of the data. In this model, the reaction rate is related to the fraction of the solid oxide which remains after a given reaction time. No essential difference was found in the fluorination reaction with the two samples of uranium oxide. An average activation energy of 30 kcal/mole was calculated for the fluorination reaction. The value of the activation energy obtained in these experiments is considered to be a more reliable value than the preliminary value of 75 kcal/mole obtained in previous studies (see ANL-6619).

d. Heat Transfer Studies: U_3O_8 -Alundum Fluidized-bed Systems. Heat transfer studies of uranium oxide (U_3O_8) fines in fluidized-bed systems were made in a 3-in.-diameter glass column. Uranium oxide (U_3O_8) fines of about 4- μ mean diameter and Alundum of 60 to 170 mesh size were used in these studies. A cylindrical heater with attached thermocouples served as an internal probe for the heat transfer determinations.

Heat transfer was improved in a bed of U_3O_8 fines (no Alundum present) by superimposing a pulsed gas flow on a steady flow of fluidizing gas. Both the pulsing gas and fluidizing gas were air. Channeling in the U_3O_8 fluid-bed column was substantially eliminated by gas pulsing. Although pulsing improved heat transfer in the fluid bed at superficial gas velocities greater than 0.2 ft/sec, the rate of pulsing did not have a significant effect on the heat transfer coefficient from the surface of an internal heater to the bed of U_3O_8 fines. At the lower superficial gas velocities (less than 0.2 ft/sec), high pulse rates (greater than 25 pulses/min) provided better heat transfer. Without gas pulsing, heat transfer coefficients could not be reproduced in a bed of U_3O_8 fines (no Alundum present), because of severe channeling.

In heat transfer studies of mixtures of U_3O_8 fines and Alundum at gas pulse rates of about 20 pulses/min, small increases in the heat transfer coefficient were observed upon increasing the Alundum content from 0 to 100 w/o and upon increasing the superficial gas velocity from 0.1 to 0.9 ft/sec. The effect of the superficial gas velocity on the heat transfer coefficient was less at the higher Alundum compositions in the U_3O_8 -Alundum mixtures. The data indicate a maximum value for the heat transfer coefficient at about 15 w/o U_3O_8 -85 w/o Alundum and a superficial gas velocity of 0.6 ft/sec.

In heat transfer studies made without gas pulsing, heat transfer coefficients of mixtures containing from 65 to 100 w/o Alundum and from 35 to zero w/o U_3O_8 fines did not differ significantly from the coefficients obtained with similar mixtures which were subjected to gas pulsing.

e. Decladding of Fuel Elements by Fluid-bed Oxidation. The development of a fluid-bed oxidation method for the separation of uranium dioxide pellets from stainless steel cladding in typical fuel elements is continuing (see Progress Report for January 1963, ANL-6683, p. 42). In this method, the uranium dioxide is oxidized with air to a higher oxide (U_3O_8). The uranium dioxide pellets break into fragments because of the large internal stresses produced by the difference in densities of the two oxides (UO_2 and U_3O_8). The U_3O_8 fines produced in the oxidation zone of the two-zone oxidation-fluorination reactor are elutriated to the fluid-bed fluorination zone.

Preliminary laboratory experiments were carried out in a fluid-bed reactor to evaluate the oxidative decladding method for the removal of uranium dioxide pellets from stainless steel cladding which had been slotted longitudinally at regular intervals. The slotting procedure, which was developed at Atomics International, is helpful in rupturing the cladding and in allowing greater access of the oxidizing gas to the UO_2 pellets. The simulated fuel elements used in this study were made by placing twelve $\frac{1}{2}$ -in.-diameter uranium dioxide pellets in $5\frac{1}{2}$ -in.-long stainless steel tubes having a 10-mil wall thickness. Slots ($\frac{1}{8}$ in. by $\frac{1}{32}$ in.) were milled lengthwise in the stainless steel tubes at intervals of $\frac{3}{8}$ in. The results of a run made with the simulated fuel elements in a fluidized bed of alumina have shown that complete splitting of the cladding and a complete removal of uranium dioxide pellets can be accomplished by oxidation with air in 4 hr at $450^\circ C$. The split cladding appeared free of any uranium oxide. The U_3O_8 fines formed in the oxidation reaction mixed with the alumina bed material in the fluid-bed reactor.

Preliminary engineering-scale runs have shown that a high reaction temperature ($450^\circ C$) and gas pulsing improve the rate at which uranium dioxide pellets are separated from stainless steel cladding in a bed of

$1\frac{1}{4}$ -in.-long simulated fuel elements. In a similar run, perforating the cladding with one hole (of $\frac{7}{64}$ -in. diameter) did not affect the rate at which uranium dioxide pellets are separated from the stainless steel cladding.

f. Chlorination and Fluorination Steps in the Separation of Uranium from Aluminum Alloy Fuels. The feasibility of using the fluid-bed chlorination-fluorination scheme for the recovery of uranium from uranium-aluminum alloy fuels was studied. The chlorination-fluorination reaction sequence included hydrochlorination at 200°C for $3\frac{1}{4}$ hr, fluorination at 350°C for $1\frac{1}{2}$ hr, followed by further fluorination at 500°C for 5 hr. A charge of 240 g of 5 to 10 w/o uranium-aluminum alloy clad in aluminum was used. The charge was in the form of chips $\frac{1}{16}$ -in. thick by $\frac{1}{8}$ to $\frac{1}{4}$ in. wide by $\frac{1}{8}$ to $\frac{3}{8}$ in. long.

About 60 percent of the charge reacted with the hydrogen chloride at an average rate of 44 g/hr. During this period, the utilization efficiency for hydrogen chloride was about 90 percent. No further reaction of the residual alloy chips was noted during the two fluorination steps. The fluorine concentration at the end of the fluorination steps was about 90 percent. No significant reaction between the fluorine and the Alundum was noted. A microscopic examination of a number of residual alloy chips showed them to be thinner than the alloy chips in the initial charge. This suggests that hydrogen chloride reacted first with the aluminum cladding and then with the uranium-aluminum alloy.

The uranium material balance showed that about 57 percent of the uranium in the initial charge remained in the unreacted alloy chips, thereby indicating that no preferential removal of the uranium from the alloy chips had occurred. Of the initial uranium charged, about 0.8 w/o was lost through the static Alundum bed filter during the hydrochlorination step, and about 3 w/o was retained on the Alundum bed of the reactor.

g. Fluid-bed Hydrolysis of Zirconium Tetrachloride. Investigation of the fluid-bed hydrolysis (by steam) of zirconium tetrachloride to zirconium dioxide was continued (see Progress Report for November 1962, ANL-6658, p. 52). Current studies in the 6-in.-diameter reactor column were made to evaluate the effect of using higher zirconium tetrachloride feed rates (4.1 to 5.1 kg/hr) than were used previously (2 to 3 kg/hr) and to determine whether beds comprised of fine material would be operable for extended periods of time. Sand was used as the starting bed material. A bed of fine sand (96.5 percent of -100 mesh, 78 percent of -200 mesh) was prepared to simulate a bed such as might be encountered in the later stages of a long run. Four runs ranging from $2\frac{1}{4}$ - to $5\frac{3}{4}$ -hr duration were made.

The four runs were all completed satisfactorily, as indicated by the following results: (a) no pressure increase occurred across the exit gas filters; (b) conversion of the zirconium tetrachloride vapor to the solid zirconium oxide was complete (as indicated by the absence of zirconium in the off-gas condensate stream); (c) no caking of the bed occurred, even when the bed consisted primarily of fines (< 200 mesh material); and (d) good material balances (~98 percent) were obtained.

h. Conversion of Uranium Hexafluoride to Uranium Dioxide: Preparation of High-density Particles. The fluid-bed preparation of 80 to 100 mesh size dense uranium dioxide particles directly from uranium hexafluoride by the simultaneous reaction of the hexafluoride with steam and hydrogen has been investigated (see Progress Report for December 1962, ANL-6672, p. 33). Uranium dioxide particles of this size range are suitable for use in the fabrication of vibratory compacted or dispersion-type fuel elements. Uranium dioxide having the desired particle mesh size (80 to 100) and having a particle density of 9.4 g/cc (measured by mercury displacement) was prepared from a starting bed of 100 to 140 mesh uranium dioxide particles. The uranium dioxide product was composed of 62 percent of +100 mesh material and less than one percent of +80 mesh material.

3. Calorimetry

The following thermodynamic data at 25°C have been calculated for the formation of zinc fluoride by the combination of zinc in fluorine: (1) energy of formation, $\Delta E_f^\circ = \Delta E_c^\circ = -182.4 \pm 0.4$ kcal/mole; (2) heat of formation, $\Delta H_f^\circ = -183.0 \pm 0.4$ kcal/mole; (3) Gibbs energy of formation, $\Delta G_f^\circ = -170.8 \pm 0.4$ kcal/mole. The entropy of formation, ΔS_f° , was calculated to be -40.8 cal/(deg)(mole). The heat of formation obtained by fluorine bomb calorimetry, -183.0 kcal/mole, can be compared with the value of -182.9 kcal/mole obtained from an equilibrium study.²

²L. Domange, Ann. Chim., 7, 225 (1937).

IV. PLUTONIUM RECYCLE PROGRAM

A. Core Design

The irradiation of an experimental core in the EBWR is part of the program planned for the Plutonium Recycle study. A three-zone core is being designed and will consist of 36 plutonium fuel elements (fuel composition: 2.5% PuO_2 -97.5% U^{238}O_2) in the central zone, which is surrounded by a single row of enriched uranium fuel elements to form the shim or driver zone, and a third zone of natural uranium fuel elements to form the blanket. The shim zone will be replaced periodically to provide the necessary reactivity for long (burnup) irradiations.

A study has been made of the use of a burnable poison in the shim zone to extend its life and limit the range of excess reactivity present over the period of irradiation.

Calculations have been made on the effect of an addition of europium and samarium in the atomic ratio of 1 part of each to 1000 parts of U^{235} in a shim zone enriched to 6%. It appears that one shim loading containing burnable poison can provide sufficient reactivity to reach the burnup provided by two successive unpoisoned shim loads which would normally be enriched up to 5% U^{235} . In addition, the reactivity swing is limited to a change of only 2 to 3% over the irradiation period.

B. Hydrodynamic Performance

The hydrodynamic behavior of the proposed three-zone core is being determined by iterative calculations with the use of the PDQ code to provide the flux and power distribution. The calculations were carried out by the RECHOP IBM-704 code to determine the average void fractions. Experimental data from the EBWR Core-1A experiments on carryunder and carryover were included. Based on these data, a second PDQ calculation of neutron flux was made and a second iterative hydrodynamic calculation has been initiated.

A new computer code (CHOPPED) has been constructed to solve the hydrodynamics equations. The new code differs from RECHOP by the use of power distribution as input and produces the void fraction profile in a channel. This code will simplify the iteration procedure for boiling water cores, and it should be possible to obtain reasonably accurate graphical representation of the average void fraction in each zone and the total reactor power. The change will also permit the PDQ calculations to be performed independently over a power range from 20 to 100 Mw.

C. Fuel Development

The responsibility for providing fuel for the Plutonium Recycle Experiment is divided between G. E. Hanford and ANL. Hanford is responsible for fabricating all plutonium fuel for the core and ANL is responsible for procuring all the uranium fuel for the driver and shim zone of the core.

The vibratory compaction of mixed PuO_2 - UO_2 powders has been developed at Hanford for loading the fuel into Zircaloy-2 fuel tubes. Drawings are complete for the removable type plutonium fuel rods and orders have been placed for the Zircaloy-2 fuel-rod-jacket tubing.

A tentative selection of 0.046 w/o Eu_2O_3 + 0.077 w/o Sm_2O_3 mixed rare earth oxide in UO_2 with an isotopic enrichment of 6.0 w/o U^{235} has been made for the first shim loading. Drawings and specifications are complete for the uranium fuel procurement, and the bid invitation package has been prepared.

V. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Core Physics

An IBM-704 code (1559/RE) has been written to solve the infinite medium transport equation in the presence of: (a) isotropic homogeneous scattering; and (b) scattering, including Doppler broadening effects, and with absorption by (up to) four heavy elements, each with (up to) 75 single-level Breit-Wigner resonances. It is further assumed that the average logarithmic decrement in neutron energy after any heavy atom scattering is a constant for each problem.

The code will be used to establish the Doppler coefficients in the resolved resonance region of AARR, and to investigate resonance interference in heavily loaded cores, such as used in the Yankee Pressurized Water Power Reactor and the Hi-C critical experiments in ZPR-VII.

The computer time required for the solution of these problems is relatively long and can be determined from the following equation:

$$\text{Running Time} = \left(\frac{\Delta U \times NL \times N}{A} \right) 5.7 \times 10^{-5} \text{ min} ,$$

where

ΔU = total lethargy width;

NL = total number of levels;

N = number of subintervals per heavy atom logarithmic energy decrement;

A = average heavy atom scattering mass.

2. Reactor Control Studies

By use of a slightly modified version of the method of analysis presented by S. S. L. Chang, Synthesis of Optimum Control Systems, McGraw-Hill Book Co., Inc., New York (1961), hand calculations were performed to determine optimum control ratios in the sense that the integral-square system error was constrained and the integral-square acceleration of the control rod was minimized. Calculations were based on an approximate version of the reactor transfer function (with inherent feedback) at 100 Mw. These preliminary results indicate that the controlled reactor system is characterized by a pair of dominant complex roots with a damping ratio of approximately 0.7 which remains essentially constant as the

integral-square error constraint is varied. These promising results indicate the desirability of extending the calculations to other power levels using a more accurate representation of the reactor transfer function.

Some literature on the design of reactivity computers was obtained and is being studied to determine possible application for use in a predictive control loop. The analog circuits described by G. S. Stubbs³ appear promising in this respect.

3. Primary System Design - Particulate Separators

The AARR primary system design as presented in the United Nuclear Corporation Preliminary Feasibility and Cost Study includes two settling tanks, one in each of the 16-in. outlet lines. The purpose of these pieces of equipment is to remove particulate radioactive material, which could be formed in the event of an accidental fuel meltdown, from the circulating coolant before they are carried further into the heat exchangers, pumps, and piping of the primary circuit. In this manner it is hoped to confine as much of the highly radioactive debris as possible to a location where it would be easily removed from the system. If such material were deposited in the heat exchangers and other equipment after a fuel meltdown, decontamination would be a very difficult task.

A rough analysis of the settling tanks described in the UNC report, based on the design flow rate and settling velocities for particles of density similar to that of the core material, indicates that the smallest particle which these tanks could be expected to remove would be about 4 mm (0.16 in.) in diameter. Inasmuch as a fuel meltdown would very conceivably produce a considerable quantity of material of particle size smaller than 4 mm, it is proposed to replace the settling tanks with centrifugal particle separators. Limited tests with a plastic model using a linear flow rate in the inlet pipe equal to that in the 16-in. outlet of AARR at design flow rate indicate that this type of equipment can be designed to remove very small particles and still not introduce a prohibitive pressure drop into the system.

Discussion with a manufacturer of such equipment has indicated that for AARR conditions, eight parallel cyclone-type separators, each 3 ft in diameter and 15 ft high, would be required. Four of these would be placed in each of the primary outlet lines from the reactor vessel. This equipment has been substituted for the previously specified settling tanks in the reference facility design. It is anticipated, however, that a further evaluation will be made by the architect-engineer when Title I design is undertaken, before a final decision is made.

³ G. S. Stubbs, IRE Transactions of the Professional Group on Nuclear Science, Vol. NS-4, No. 1 (March, 1957).

B. Direct Conversion Studies

1. Magnetohydrodynamic Systems

In magnetohydrodynamic (MHD) systems, the generation of electrical energy from a moving fluid is expressed by the following vector relationship:

$$E = B \times V \quad ,$$

where

E = voltage generated

B = magnetic field

V = velocity of the fluid.

In practice, the fluid is forced to flow through a flattened duct between the poles of a strong magnet. A voltage will be generated in the fluid at right angles to the magnetic field and the direction of flow. If electrodes are placed in this plane, a current will flow.

It is necessary that the fluid be a good electrical conductor, so that either ionized gas, plasma, or liquid metals may be used as the fluid.

A study is in progress to determine the feasibility of using a liquid metal as the fluid, and a number of concepts are being studied. Two concepts are discussed here. Both concepts use the thermodynamic properties of the fluid to convert heat into kinetic energy. Sodium is being considered as the fluid in both concepts. A MHD system has an advantage of being able to operate at reasonably high temperatures and without the use of moving parts in the conversion system. The system is well adapted for use in space with a nuclear reactor, since the fluid temperatures required can be obtained by direct heating and the fluid velocities required are in the range of current technology. The calculations indicate that reasonable conversion efficiencies can be obtained.

The first concept uses the flashing technique for conversion of the heat energy in the fluid to kinetic energy. Bulk sodium is heated in the reactor and flows into the entrance region of a nozzle. As the pressure decreases, a portion of the heated fluid flashes into vapor and is accelerated. The kinetic energy in the vapor is then transferred to the liquid particles with which it is in intimate contact. The mixed fluid is then centrifugally separated. The liquid is passed through the magnetic field to generate power, and the vapor is bypassed, condensed, and recombined with liquid for recycle through the reactor.

The injection-recirculation system uses a portion of the liquid metal as the heat absorption medium. This portion of liquid metal is vaporized at

pressure in the reactor and expanded in an injector to high velocity. Simultaneously, a second stream of liquid metal flows into the low-pressure region of the injector and mixes with the high-velocity vapor. The accelerated liquid stream is then passed between the poles of a magnet with consequent generation of power. The liquid then flows into an expanding duct to decrease its velocity and reduce friction losses. A portion of the liquid is bypassed and directed back to the energy source for reheating. The main stream, still moving at an appreciable velocity, flows to a heat exchanger where it is cooled and then enters a converging duct to increase its velocity before re-entering the combining section of the injector, to repeat the cycle.

A series of calculations have been made for both cycles over the operating temperature range of 1300°C to 800°C. A preliminary comparison of the flashing and recirculating system performances based upon reasonable assumptions for nozzle, combiner, and friction losses are shown in Table XII for a system designed to produce 4 kw of electricity.

Table XII. Comparison of Performance

<u>Description</u>	<u>Type of Cycle</u>	
	<u>Flashing</u>	<u>Recirculation</u>
Fluid	Sodium	Sodium
Cycle Efficiency, %	13.1	21
Flow Rate, gm/sec	638	278.5
Velocity of Film through Magnet, cm/sec	18,600	28,200
Output, kw	4	4

The principal areas of research involve detailed investigation into the:

- (a) measurements of energy transfer between the high-velocity vapor and the liquid;
- (b) design of nozzle and combiner to provide efficient transfer of energy;
- (c) analysis of friction losses in system and development of techniques for minimizing pressure and energy losses; and
- (d) optimization of design of nuclear reactor and converter system to provide maximum overall efficiency.

Experimental apparatus has been assembled to study both the flashing and recirculating injection concepts. The apparatus for the flashing cycle, shown in Figure 9, consists of a special nozzle made of transparent

Lucite which is placed between the poles of a strong electromagnet. Sodium-potassium was selected as the liquid metal fluid and nitrogen gas was substituted for the metal vapor to eliminate the need for heating.

Figure 9. Experimental Apparatus Used to Study the Flashing Cycle Concept of Electrical Generation. The photograph shows the Lucite nozzle positioned between the pole faces of the electromagnet.

The apparatus for the injector cycle, shown in Figure 10, uses steam and water for the vapor and liquid, respectively. A flow diagram of this system is shown in Figure 11. The system is now being tested and calibrated prior to initiating the experimental program.

Figure 10
Experimental Apparatus Used to Study the Injector Recirculating Cycle Concept of Electrical Generation.

Figure 11. Schematic Layout of Apparatus Used to Study the Injector Recirculating Cycle Concept.

2. The Cesium Diode

A theory has been developed for the behavior of a steady-state ion-rich plasma in the cesium diode. It includes the effect of sheath voltages on the output current versus output voltage characteristics. It improves on older treatments of the subject⁴ by taking a more accurate account of the ion currents produced by resonance ionization on the emitter surface from the arriving current of neutral cesium atoms.

When the collector of the cesium diode is made negative enough with respect to the electron emitter so that almost none of the electrons reach the collector, the collector current is composed almost entirely of ions formed on the emitter surface. The bias voltage necessary to establish this condition with an ion-rich plasma is about -5 v, providing other processes are not contributing significantly to the current. Sources which can add to the ion current from the emitter and complicate the process are photoelectrons or thermionic electrons from the collector, and ions produced in the plasma volume from the excited atoms or ground state atoms.

The expression for the ion current density in ma/cm² observed at the collector is

$$J = \left[\frac{j_{\text{sat}} J_{\text{sat}}}{2G} \right]^{1/2},$$

where

$$j_{\text{sat}} = AT_E^2 \exp\left(-\frac{e\phi_E}{kT_E}\right) = \text{electron current density from the emitter}$$

T_E = emitter temperature

A = the Richardson constant

e = electronic charge

ϕ_E = emitter work function

k = Boltzmann constant

J_{sat} = resonance ion current density at the emitter surface

G = ratio of electron velocity to the cesium ion velocity in the plasma, 494.

⁴E. N. Carabateas, S. D. Pezaris, and G. N. Hatsopoulos, J. Appl. Phys., 32, 352 (1961).

This formula can be used to check the experimental data shown in ANL-6610 (Monthly Progress Report for August, 1962, Figure 17, p. 64). In this case, the plasma is ion rich from resonance ionization at the emitter, and conditions are far from those producing the low-voltage arc mode, according to the data for tantalum given by Nottingham.⁵ Other sources which can add to the ion current from the emitter are probably negligible in this case as long as the bias voltage is below -5 or -10 v. Using the previously determined electron emission constants,

$$A = 60.5 \times 10^3 \text{ ma/cm}^2 (\text{°K})^2 ,$$

$$\phi_E = 4.1 \pm 0.1 \text{ volt,}$$

and the given ion current density at the emitter surface, the ion currents at the collector, J , were calculated and are shown in Figure 12 as dotted lines which are superimposed on Figure 17 in ANL-6610. No arbitrary fitting parameters were used. Considering that the cesium bath temperature could not be held constant during the runs and that the uncertainty in T_E is about 50°K, the agreement is quite good.

The analysis explains how the ion current observed at the collector depends strongly on the emitter temperature even though the ion current from the emitter surface itself is almost independent of emitter temperature. The solution for the steady-state condition does not deny the existence of the oscillatory solutions; oscillatory outputs have also been experimentally observed.

3. Regenerative EMF Cells

The investigation of thermally regenerative emf cells was continued (see Progress Report for November 1962, ANL-6658, p. 63). The major endeavor was directed toward the study of bimetallic cells of the type

where M_1 and M_2 are two metals.

Studies of the lithium-bismuth cell were carried out with $\text{Li}_3\text{-Bi}$ -saturated cathodes and both LiCl-KCl and LiF-LiCl mixtures as electrolytes. Voltage instability of the cell associated with the high-temperature reduction of potassium chloride by the molten lithium metal anode was observed above 475°C. This difficulty was eliminated by the use of a LiCl-LiF salt mixture as the electrolyte. Cells with Li_3Bi -saturated cathodes

⁵ Proceedings of the Third Government-Industry Thermionic Round Table Discussions, AD-270905, Vol. I, pp. 1-13, Figure 2.

yielded stable potentials, whereas cells with unsaturated cathodes did not, perhaps because of irreversible transfer of lithium into the cathode. The open-circuit voltages for cells with Li_3Bi -saturated cathodes were found to range from 0.56 v at 831°C to 0.72 v at 486°C in the case of a cell using LiCl-LiF as the electrolyte, and from 0.71 v at 528°C to 0.74 v at 432°C for a cell using LiCl-KCl (60 m/o Li) as the electrolyte.

Figure 12. The Curves Show the Experimental and Calculated Relationships Between the Emitter Temperature, Radiofrequency Output, and the Ion Current Density to the Collector for Various Cesium Temperatures, T_{Cs} . The tests were conducted with -5 to -10 v being applied to the collector with the emitter grounded.

In support of the regenerative emf cell work, phase studies are being made on salt mixtures that may be used as electrolytes in the cells. The phase diagram of the lithium hydride-lithium chloride system is nearly complete. A eutectic, which melts at 496°C , was found at 34 m/o lithium hydride.

VI. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal Oxidation and Ignition Studies

The rates of oxidation of uranium by air have been measured over the range from 600 to 1000°C. The experiments were carried out by passing a mixture of air and argon over one-cm cubes of uranium which were supported on thermocouples and heated to the desired temperature by an induction coil. The depletion of oxygen and nitrogen in the effluent gas was measured continuously by a mass spectrometer.

Results of these experiments have shown that at 600 and 700°C the rates of the oxidation reactions are nearly linear. At 900 and 1000°C, the reaction rates appear to be parabolic initially, and then become linear as the oxidation progresses. At 800°C, the rate is very nearly linear and falls between the rate at 700°C and the overall rate at 900°C. No evidence of nitrogen reaction was found over the temperature range studied.

2. Metal-Water Studies

Reaction of stainless steel-302 with water was studied by means of the condenser-discharge method. In this method, samples in the form of wires are heated in a water environment by a surge current from a bank of condensers. The energy input to the wires is measured electrically and used to calculate the initial reaction temperature. The present studies were carried out with 30-mil, one-inch-long wires over an initial temperature range from 2000 to 3000°C. Both room-temperature water and water heated to 200°C were used in the experiments. In room-temperature water, 6 and 14 percent of the stainless steel-302 wires reacted at initial reaction temperatures of about 2000 and 3000°C, respectively. In water at 200°C, 12.5 and 34.0 percent of the metal reacted at about 2000 and 3000°C, respectively. The results obtained in these experiments are in good agreement with the results obtained in experiments with stainless steel-316 (see ANL-6596, p.186).

In-pile studies of metal-water reactions in the TREAT facility were continued. Three experiments were carried out with unclad 81 w/o aluminum-17 w/o uranium (fully enriched)-2 w/o nickel alloy plates. The energy input to the plates was calculated to be 473 cal/g of plate in one run and 530 cal/g in the other two runs. The extent of the aluminum-water reaction ranged from 4.5 to 14.2 percent. Each plate was transformed to a single globule of metal. Metallographic examination of the globules indicated that the porosity of the metal increased with the extent of the aluminum-water reaction.

B. Fast Reactor Safety Studies

1. Meltdown Experiments

Fast power reactor designs characteristically contain sufficient fuel to produce a super-prompt critical assembly if the core were rearranged into a sufficiently compact mass. Hence, it is possible to postulate a class of "meltdown" incidents in which the core is subjected to abnormal operation, some degree of fuel failure occurs, and the fuel moves into a more compact mass to produce a destructive burst of nuclear power. TREAT is being used to perform in-pile experiments to study the causes and characteristics of failure of fast reactor fuel elements and to survey the mechanisms influencing movement of fuel.

a. In-pile Tests on Time of Failure of Type 304 Stainless Steel-clad Fuel Pins. A previous laboratory investigation of the penetration of both uranium and uranium-5 w/o fissium alloy through iron and Type 304 stainless steel was made by a dip method in the temperature range from 1100°C to 1350°C.⁶ The TREAT experiments more closely duplicate actual reactor conditions, with respect to sample geometry, and with respect to time and space variations of temperature. The isothermal dip experiments showed that the rates of penetration of iron and stainless steel decreased with increasing temperature over a portion of the temperature range studied. These results were confirmed by experiment.

A series of experiments to measure times of sample failure were performed in TREAT under conditions designed to minimize error and uncertainties with respect to energy inputs into the sample. Two half-length fuel sample elements, one with and one without a sodium bond, were tested in a modified opaque capsule. When subjected to a power burst in TREAT, the uranium-5 w/o fissium fuel alloy melted and penetrated its stainless steel jacket. This penetration completed an electrical circuit to a steel sheath surrounding the element. The electrical signal was indicated on a high-speed recorder along with the output signal from the high-speed thermocouples attached to the sample jacket. The penetration time could be deduced from the data.

The energy input into the sample was determined by programming the initial reactivity input. The power excursion was terminated by the reactor prompt negative temperature coefficient of reactivity. A temperature range from approximately 1100°C to 1250°C was investigated.

The unbonded samples gave erratic results.

⁶C. M. Walter and L. R. Kelman, Penetration Rate Studies of Stainless Steel by Molten-uranium and Uranium-Fissium Alloy, J. Nuclear Materials, 6, 281-290 (1962).

For the modest transients, penetration times for the sodium-bonded elements (9-mil cladding) were found to be about 2 sec. An intermediate transient produced a penetration time of about 0.7 sec. Penetration time for a transient near the upper end of the temperature range was about 1.1 sec. The bonded sample exposed to the most severe transient failed almost instantly, but previous calculations indicated that failure could have resulted from bursting of the steel cladding due to internal pressure. Results of the more difficult and time-consuming in-pile tests are in reasonable agreement with the laboratory-dip experiments for uranium-5 w/o fission alloy fuel and stainless steel cladding. A similar series of TREAT tests is being conducted on stainless steel-clad uranium pins.

b. Transparent Facility for Photographic Experiments with Pre-irradiated Samples. Equipment has been designed for use in photographic meltdown experiments on pre-irradiated specimens. The equipment permits direct observation of the pre-failure sample conditions, phenomena occurring at failure, and the form and velocities of material leaving sample fuel elements on failure. The transparent facility primarily consists of the sample, a test capsule, slot liner, and a camera and lens system. The principal difference between this and the opaque system is in the use of fused silica windows in the capsule and slot liner, which provides a path for the light from the sample to the camera.

The test capsule (a double-walled Zircaloy container fitted with a high-purity fused silica window) holds the test specimen and is not sealed. The capsule provides support for the specimen, primary containment for meltdown products, and affords capability for pressure relief in the event that that feature is necessary. The cover gas (helium) flows first into the capsule, thence into the slot-liner subassembly which contains the capsule. The helium cover gas is then exhausted through cooled charcoal filters to the reactor stack. Capsule parts have been designed to permit remote encapsulation of samples in the cave. However, the capsule and window are designed to be interchangeable with parts from the opaque system. All capsule parts have been fabricated. Figure 13 shows a capsule in the remote assembly fixture.

The slot liner is a stainless steel box fitted with a high-purity fused silica window. This subassembly holds the test capsule in position in the reactor test slot which extends from the graphite reflector into the core center. The slot liner has pressure-containment capability and is lined on the inside with graphite and an inner Zircaloy sleeve. The graphite provides for protection against attack from meltdown products. The Zircaloy liner is designed to contain gaseous or particulate contamination, and can be removed easily for decontamination. One prototype slot liner has been fabricated and leak tested. Although the test pressure for transparent slot liners was set at 0.4 atm, the prototype liner was tested to 0.67 atm. At the conclusion of the test, the liner was checked and found to have no permanent deformation due to pressurizing. Although the new slot liner is different from the old, because of changes caused by necessity for remote

assembly and disassembly, it can be handled with the same TREAT equipment as the standard one for non-radioactive samples. Figure 14 shows the new liner, sans window, and flanges.

Figure 13. Test Capsule Prototype is Shown Mounted on the Remote Assembly Fixture in the Cave Mockup in Building 206. Part A is the capsule proper. Part B is the sample load frame which is inserted into the capsule. The frame holds the sample while thermocouples are welded to the cladding, and is used to load the instrumented specimen into the capsule and hold the sample in the capsule. Part C holds the capsule window and is used to slide it into position after the load frame is in place. The sample shown (D) is an EBR-II element. Clamps (E) are used to weld thermocouples to the sample.

Figure 14

Slot Liner Prototype is Shown, with Window Flanges and Window Removed. The end of the Zircaloy sleeve has been slid out about 7 cm from the end of the liner.

The shipping coffin is a dual, sealed coffin designed to transport two liners. It is being fabricated by a subcontractor.

Remote assembly procedures and equipment have been checked in a cave mockup in Bldg. 206. The mockup includes two manipulators and a simulated cave window, so that design and handling can be studied in detail. Placement of coffin, liner, capsule, and capsule-assembly equipment were checked in the cave facility earlier, with full-size wooden models of the coffin and slot liner.

2. Installation of Sodium Loop in TREAT

The three defective, 4-in., cylinder-operated valves for the loop (see Progress Report for December 1962, ANL-6672, p. 44) were returned to the manufacturer for repair and operator modifications. The two valves of the same type which passed the initial inspection were also returned for modification of the operators.

Installation of the conduit between the control panels and the main floor of the Reactor Building and Sodium Equipment Room was completed. Work continued on the installation of wiring between instrument and electrical control panels.

The contractor completed installation of the major equipment items for the Sodium Equipment Room air-cooling system and installation of piping between units is in progress.

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AND PRESSURES FROM 1 TO 50 ATMOSPHERES
Charles E. Cohn and Gerald H. Golden