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REACTOR DEVELOPMENT PROGRAM
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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:

April 1963	ANL-6717
May 1963	ANL-6739
June 1963	ANL-6749
July 1963	ANL-6764
August 1963	ANL-6780
September 1963	ANL-6784

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I. BOILING WATER REACTORS

A. BORAX-V

1. Operations

The reactor with central superheater core (CSH-1B) was operated at power, producing superheated steam for the first time on October 10. This was the first production of superheated steam in an integral boiling water-nuclear superheating reactor.

In the 7 days of operation thus far completed, a maximum power of 10.2 MWt was reached. The integrated power during heating with the superheater flooded was 7.3 MW-hr, and the integrated power with steam flowing through the superheater was $47 \frac{3}{4}$ MW-hr during 15 hr of operating time. A summary of temperatures and steam-line radioactivity at various powers is given in Table I.

Table I. Initial Power Operation of Core CSH-1B, BORAX-V

	Power (MWt)		
	5.2	7.3	10.2
Saturated Steam Temp, °F	487	487	485
First-pass Fuel Assembly, Exit Steam Temp, °F	632	635	640
Second-pass Fuel Assembly, Exit Steam Temp, °F	755	760	760
Steam-line Temp, °F	640	660	725
Maximum Second-pass Fuel Temp, °F	785	790	820
Steam-line Radioactivity, mr/hr	50	50	75

The superheater drain, vent, and flood systems functioned satisfactorily. Draining and flooding of water in the superheater steam coolant channels were accomplished ten times with the reactor at about 600 psig. By venting steam during the period of time (approximately 50 min) required to regain operating pressure after draining, the superheater fuel temperatures were maintained below 1050°F. Because of the low amount of integrated power produced thus far, very little venting has been required during shutdown. Superheater fuel temperatures have lowered to saturation temperatures within a few minutes after reactor shutdown.

Bringing the reactor to operating temperatures and pressure by nuclear heating has been accomplished five times. At the established maximum heating power of 0.8 MWt with the superheater flooded, typical boiling-noise variations in linear flux level were noted. These variations were greater than those observed at similar power levels during operation of

boiling core B-2, probably due to water boiling in the channels of the superheater fuel coolant. With the reactor power at 7.3 MWt, the variations in linear flux level as measured on a light beam galvanometer-type oscillograph were $\pm 3.5\%$ of the average power.

During the operation at about 7 MWt, the fission product monitor for the superheated steam indicated a low-level increase in radioactivity. The monitor for saturated steam from the reactor vessel steam dome, however, indicated no increase in activity. Analysis of the anion-exchange column from the superheated-steam fission product monitor indicated the presence of the fission product I^{135} .

After the 10.2-MWt run, the reactor was shut down. A leak test, run at room-temperature conditions, indicated that the in-leakage of moderator water into the superheated steam channels had increased from the 1.5 gal/hr measured before power operation to about 50 gal/hr. Leakage of water or saturated steam had been suspected because of the large difference between second-pass exit and steam-line temperatures (see Table I). Because of the fission product and leakage problems, power operation was interrupted, and the reactor vessel head was removed to permit examination of the superheater fuel.

a. Fission Product Experiments. To determine which superheater fuel assemblies were yielding fission products, equipment was installed to obtain water samples from all twelve assemblies at a point about 4 in. above the top of the fuel plates. Water was sampled prior to a low-power run; the reactor was then operated at 100 kW for 55 min. Another water sample was then taken immediately after the run. The samples before and after the power run were counted and compared. Two tests were made with essentially the same results.

The results of the second test, showing the gross gamma count on 4-ml aliquots of each sample and counts on radiochemical iodine separations are tabulated in Table II.

Table II. Fission Product Experiment with Core CSH/1B, BORAX-V

Superheater Fuel Assembly No.	Core Position	Gross Gamma Count (cpm)	Iodine Separations Count (cpm)	Superheater Fuel Assembly No.	Core Position	Gross Gamma Count (cpm)	Iodine Separations Count (cpm)
<u>First Pass</u>				<u>Second Pass</u>			
C-1	35	45,500	2,450	C-2	34	28,500	150*
C-14-I	45	4,000	170*	C-12	43	28,800	-
C-10	46	30,400	95*	C-16-I	44	20,100	-
C-9	55	30,500	150*	C-3	53	80,000	3,860
C-11	56	38,800	53**	C-4	54	32,900	1,530
C-13	65	33,200	310*	C-6	64	28,000	140*
				Superheater Drain		2,250	295*
				Bottom of Reactor Vessel		40	-
				Average Low Count		-	190

* Used for average low count.

** Possible I_2 separation error.

The gross gamma count was made from 1-2 hr after shutdown and the values have been corrected for decay. The iodine separation count reported here was run 3 days after shutdown and is not corrected for decay. A gamma-ray-spectrometer analysis shows the iodine to be mostly I^{131} at this time.

Some fission product iodine was found in each superheater fuel assembly, but assembly C-3 was the worst, followed by C-1 and C-4. The ratio of the I_2 separation count to the average low count for these assemblies is as follows:

Assembly No.	Count
	Average Low Count
C-3	22.7
C-1	9.0
C-4	6.7

b. Superheater Fuel Inspection. A pneumatic leak test was run on superheater fuel assemblies while still in place in the reactor. Bubbles rose from between two assemblies only. Removal and individual leak testing of all three fuel assemblies in the west quadrant of the superheater indicated a leak in fuel assembly C-3 from core position 53 only. The leak was a crack about $\frac{1}{2}$ -in. long in the weld between the bottom nozzle and the 5-element fuel subassembly. Underwater visual inspection of this assembly revealed two localized bulges near the end of each of two fuel-element-insulating tubes adjacent to fillet welds made during repair of the superheat fuel assemblies. In addition, two outside and one inside fuel elements of the same assembly appeared to have a slight longitudinal bow. The internal contact points of the spacer wires on the outer insulating tubes are visible, but no undulations could be felt with a probe moved over the surface. As discussed, fuel assembly C-3 was also found to produce the major portion of the observed fission products.

The other two fuel assemblies inspected did not leak and had no visible damage. Superheater fuel assembly C-3 has been replaced with a spare assembly and has been shipped to Argonne, Illinois, for dissection and examination.

The reactor was reloaded. Another leak test at room-temperature conditions indicated an inleakage of moderator water to the superheater of 1.2 gal/hr. The fission products-detection experiment was repeated. The iodine separation counts from samples from superheater fuel assemblies C-1 and C-4 gave about the same ratio to the average low count as on the previous experiment. The count on the sample from the fresh fuel assembly, C-7, was about the same as the average low count.

This trace of iodine in a fresh assembly probably comes from either fuel-surface contamination or diffusion through the water in the interpass plenum chambers from other fuel assemblies. Preparations for resumption of power operation are underway.

2. Water Chemistry

The principal water-chemistry measurements taken during the initial power operation of central superheater core CSH-1B are given in Table III.

Table III. Initial Water-chemistry Measurements at Power
(Core CSH-1B of BORAX-V)

	Reactor Water	Saturated Steam	Superheated Steam
Resistivity, megohm-cm	2.0-2.8	0.9-1.2	1.1-1.6
pH	6.5-7.0	5.2-5.6	5.9-6.2
Cl ⁻ , ppm	0.02-0.04	<0.04	-
Suspended Solids, Average ppm	0.13	-	-
Gross specific gamma activity, c/min/ml	9,100	3,360	3,500

The high gross specific gamma activity was due to Na²⁴ which passed into the reactor with some poor-quality makeup water. No fission products were detected in either reactor water or saturated steam. The saturated steam was checked for NO₂⁻, NO₃⁻, Cl⁻, NH₄⁺, and iron ions to find the reason for the low resistivity and pH, but none were present. Elution of the anion resin on the superheated-steam fission product monitor showed the presence of CO₃⁼. This probably caused the low readings obtained from saturated steam.

As indicated by the pH and resistivity values, the superheated steam appeared to contain fewer impurities than the saturated steam. This implies that the superheater was removing these impurities. An attempt to confirm the effect by measuring the gross specific gamma activity of condensed inlet and outlet steam samples failed, probably because fission products were in the superheated steam. The process most probably responsible for the observed increase in pH and resistivity values is the reduction of CO₂ to CO on the surfaces of the superheater fuel plates.

3. Modification and Maintenance

The new hydrotest pump was utilized to perform a successful 900-psig hydrostatic test prior to power operation.

Modification of the control for the boron-addition system was completed. This change provides a 10-min time delay between the pressurizing of the boron-addition tank and the opening of the valves to deliver the boric acid solution to the reactor vessel to prevent water hammer in the tank and piping. In emergency conditions, this delay can be bypassed. This modification eliminates the need for maintaining the boron-addition system at the same pressure and temperature as the reactor vessel, a situation which has caused excessive maintenance and maloperation of some equipment.

4. Development of Advanced Superheater Fuel

An air-flow pressure-drop test was made on an advanced superheater fuel mockup containing spherical pellets packed in a cubic lattice. This mockup consists of a 24-in.-long bed of 1,536 half-in. -dia pellets contained inside a double-walled, 3-in. -ID, square tube and outside a double-walled, 1-in. -OD, square tube. Moderator water fills the center tube and surrounds the assembly.

Testing was done with an entrance pressure of 50 psig at an air temperature of 65°F. Two runs were made, as follows:

	<u>Q(lb/hr)</u>	<u>Δp(psi)</u>
Run I	2,140	37.6
Run II	2,670	47.6

Here again, as in the previous test on a rhombohedrally packed spherical pellet configuration, the pressure drop is excessive (flow rate per superheater fuel assembly equivalent to 20 MWt reactor power is about 9,000 lb/hr).

Because of this prohibitive pressure drop, the spherical pellet and pea-in-pod advanced superheater fuel configurations are deemed not feasible. Design, development, and testing on this concept has been terminated.

A design study on a superheater fuel assembly containing fuel rods having a roughened surface to enhance heat transfer has been initiated.

II. LIQUID-METAL-COOLED REACTORS

A. General Fast Reactor Physics

1. ZPR-III

a. RAPSODIE. The preliminary loading of the RAPSODIE physics experiment, Assembly 44, was completed. The loaded volume was 38.77 liters, and the experimental critical volume was 38.6 liters. The core is constructed of one-drawer unit cells. The composition and dimensions of the experiment are shown in Table IV, and Figures 1 and 2.

Table IV. RAPSODIE Composition

Zone	Mass in gm-cm ⁻³								
	Pu	U ²³⁵	U ²³⁸	Al	Fe	O	SS	Mo	Ni
I - Core	0.6509	1.9651	1.3297	0.46082	0.66949	0.5198	0.89613	-	-
II - Blanket	-	0.0179	8.548	0.3436	-	-	1.1015	1.037	-
III - Blanket	-	0.024	11.766	0.310	-	-	0.5735	-	-
IV - Blanket	-	0.026	12.401	0.312	-	-	0.5735	-	-

Figure 1. Initial Loading of RAPSODIE Experiment, Assembly 44

Figure 2
RAPSODIE Physics Critical
(Average Dimensions)

Subcritical methods of determining the shape of the curves for control rod worth were evaluated, and the worth of a portion of the control rod motion was calibrated by period measurements.

Because of the spontaneous neutron production in Pu^{240} , the reactor must be slightly subcritical for level operation at low powers. For operating powers around 1-2 W, level power traces are obtained for control rod positions which are about 1-hour subcritical from the critical rod positions.

Two blanket-loaded control rods and one core-loaded control rod were calibrated. The blanket rods, located at the core edge, were found to have a worth of 17 lh/in. over the linear portions of the curve for rod worth. For the core-loaded rod, located in one of the outer core drawers, a worth of 55 lh/in. was obtained.

Subcritical multiplication measurements, made with and without the external ZPR-III sources, showed that the neutron source from the spontaneous fissions in the plutonium has an effective strength of about 40% that of the external sources in the blanket. This is with 25 kg of plutonium in the core and an external source strength of 15 C of Po-Be (about 4×10^7 n/sec). Some modifications in instrumentation have been made to

reduce the sensitivity of the power-recording chambers and to permit safe operation at higher powers. For period measurements, peak powers on the order of 150 W may be necessary.

b. Measurements of the Doppler Coefficient of U^{238} . Analysis of experimental results has been completed for the Doppler experiments performed with the ZPR-III assemblies 43 and 43A. The details of the experiment are described in ANL-6764 (Progress Report for July 1963, p. 8) and ANL-6780 (Progress Report for August 1963, p. 7). Comparisons with calculated values have been made by one-dimensional diffusion theory with the 22-group cross-section set 763, derived from standard set 215. The compositions of the assemblies are given in Table V.

Table V. Composition of Assemblies 43 and 43A, atoms/cm³ x 10⁻²⁴

		Assembly 43	Assembly 43A
Central Zone	U^{235}	0.00114	0.00114
	U^{238}	0.00732	0.00732
	R = 24.4 cm	Carbon	0.00833
	H = 86.4 cm	Sodium	0.01061
		Stainless Steel	0.01657
Filter	U^{238}	0.02172	0.01448
	R = 31.4 cm	Carbon	0.02916
	H = 86.4 cm		
Driver	U^{235}	0.00467	0.00467
	R = 45.5 cm	U^{238}	0.00032
	= 44.7 cm (43A)	Carbon	0.03333
	H = 86.4 cm	Stainless Steel	0.03325

The experimental results for the two assemblies for heating the special Doppler fuel element, from 300 to 800°K, and the calculated values are shown in Table VI. The figures are for the total reactivity change on heating the element.

Table VI. Change in Reactivity in the Assemblies Due to Heating

Assembly	Observed $\Delta k/k$	Calculated $\Delta k/k$
43	-0.905×10^{-5}	-1.002×10^{-5}
43A	-1.538×10^{-5}	-2.221×10^{-5}

One measurement was made over the temperature range 300-553°K in Assembly 43A. The experimental result for this case was $-0.787 \times 10^{-5} \Delta k/k$.

The above results were obtained with the core drawers adjacent to the elements and arranged so that all the sodium cans were next to the element and the remaining plates as far as possible from the element. This was done to minimize the Dancoff interaction between the U^{238} in the drawer and that in the element. One experiment was done on Assembly 43 with these drawers reversed, that is, with the drawer of U^{238} and other materials next to the element, in order to estimate the magnitude of this effect. The Doppler change of reactivity for this case was $-0.860 \times 10^{-5} \Delta k/k$, a reduction of 5%. Attempts at calculating this change have led to much higher values, of the order of 30%.

A test of non-Doppler reactivity effects of the heated sample was made on Assembly 43. Natural boron carbide plates, $\frac{1}{2}$ in. thick, were placed around the element in order to depress the low-energy flux. The measured reactivity change on heating the sample inside this boron shield was $-0.174 \times 10^{-5} \Delta k/k$. The calculated residual Doppler effect was $-0.149 \times 10^{-5} \Delta k/k$. Thus, approximately $(-0.174 \times 10^{-5}) - (-0.149 \times 10^{-5}) = -0.025 \times 10^{-5} \Delta k/k$, of the total reactivity change of $-0.905 \times 10^{-5} \Delta k/k$ represents non-Doppler reactivity effects. This is approximately a 3% effect.

Direct measurement of the worth of U^{238} movement at the core center of Assembly 43A (simulating expansion) was made with ZPR-III U^{238} plates. A simple aluminum tube which extended axially through the reactor in one of the regular element channels was loaded with plates over a length equal to half the Doppler element length, then connected to the regular element drive mechanism. One end of the loaded section was located exactly at the center of the axial dimension of the core. An identical fixed section of U^{238} plates was placed in the opposite half of the core with one end at the core center, in order to complete the simulation of the real Doppler element. The worth of movement of the loaded tube section away from the reactor center was measured. The incremental worth of Doppler element expansion as deduced from this experiment was $+0.019 \times 10^{-5} (\Delta k/k)/mm$. Free expansion of the element from 300-800°K would be about 2 mm.

2. ZPR-VI

During this reporting period, the ZPR-VI facility was operated with Assembly No. 1. Some modifications of the experimental equipment were made. The mechanical reproducibility of control rod position and closing of the two halves were also measured.

Experimental determinations of the U^{238}/U^{235} fission ratios, using solid-state detectors, gap worth, and material-replacement experiments, were performed at the edge of the core, with such materials as U^{235} ,

tungsten, and rhenium. Central reactivity coefficient measurements were made for various materials: carbon, beryllium, tantalum, sodium, zirconium, lithium, sulfur, manganese, gold, copper, and B_4C , by means of a remotely operated sample changer. The analysis of the experimental data is currently in progress.

3. ZPR-IX

The improved seal channel and solid silicone-rubber seal have been installed on the inner airlock door. The results of preliminary tests indicate a considerable improvement in sealing effectiveness.

The drawers and matrix bundles have been sent to a vendor for coating and should be returned next month.

The source drives have been checked with dummy sources, which were found to be slightly too long to pass the curve in the pot. This was corrected by expanding the channel with a swaging tool. The active sources have been delivered but will not be installed until startup in order to minimize the radiation exposure of personnel working on the reactor. All other mechanical equipment is ready for operation.

Some minor modifications are being made in the control system to improve the latching mechanism on the control rods. The associated electrical equipment is ready for operation.

A device to measure thermal expansion coefficients of reactivity has been designed. Materials are being procured for the construction of solid-state detectors - both single detectors and four-detector arrays - which are to be used to make flux traverses. A traverse mechanism is being designed.

A contract has just been let for the delivery of beryllium oxide in 5 mo. The bulk of the tungsten order has been received. The aluminum required for the reflector is expected in two to three months, although efforts are being made to advance the date. Methods for fabricating aluminum cans to contain enriched boron are under study.

B. General Fast Reactor Fuel Development

1. Metal Fuels

a. Properties of U-Pu-Fs. The mechanical properties of uranium and uranium-plutonium alloys containing 0 and 10 w/o fission products have been measured. The ultimate tensile strength and elongation vary with temperature as shown in Figure 3. Within each phase field, the elongation increases with temperature as indicated.

Figure 3. Uranium-Plutonium-Fizzium Alloys. Ultimate tensile strength vs temperature. Injection Castings.

Considerable scatter in the data occurs below 550°C for the U-10 w/o Pu-10 w/o Fz alloy. The scatter increases with decreasing temperature (the lowest temperature data are therefore not reported). Kinetics studies indicate that this scatter is related to the degree of transformation of γ to low-temperature phases and that the time at temperature was not adequately controlled for these tests. A series of specimens are being heat treated in accordance with our knowledge of kinetics to show the effects of transformation on strength.

The strength of the uranium and uranium-10 w/o plutonium alloy decreases smoothly with temperature. Scatter is nominal, and a discontinuity at the α - β transformation in the binary is consistent with the behavior in pure uranium.

Preliminary results of the compatibility of U-10 w/o Pu-10 w/o Fz with Type 304 stainless steel in the range from 550 and 650°C have been obtained. Owing to the etching characteristics of U-Pu-Fz we have been unable as yet to delineate clearly a diffusion band on the fuel side of the couples. There are, however, indications that a wide band is present in the fuel of at least some of the couples. A comparison of the diffusion penetration of this fuel into Type 304 stainless steel and into Nb-1% Zr (see Table VII) indicates that the former appears to suffer less

penetration by the fuel than does the latter. As in our previous experience with diffusion in the system U-5 w/o Fs versus 304 stainless steel, the majority of penetration appears to be into the fuel and not into the steel.

Table VII. Penetration of U-10 w/o Pu-10 w/o Fz into Type 304 Stainless Steel and Nb-1% Zr

Temp (°C)	Penetration (μ)					
	After 7 days		After 17 days		After 42 days	
	304 SS	Nb-1 Zr	304 SS	Nb-1 Zr	304 SS	Nb-1 Zr
550	2.29	3.30	3.30	5.59	4.83	7.87
600	4.83	6.35	7.11	9.65	9.65	16.00
650	11.18	12.70	16.00	28.7	22.35	76.2

2. Fuel Jacket Development

a. Alloy Development. The arc-melted button castings mentioned in the September Progress Report (ANL-6784, p. 12) have been rolled down to approximately 3-mm plate stock, with the exception of the V-50 w/o Mo alloy. The rolling of this alloy at 1050°C resulted in failure. No further attempts to roll the alloy are planned. The Nb-10 w/o Ti alloy has been cold worked approximately 40% and is now ready to be annealed. The other alloys will be rejacketed and further hot worked before cold rolling will be attempted.

b. Duplex Tubing. The extruded rod of Type 304 stainless steel with a vanadium core (see August Progress Report, ANL-6780, p. 9) was drilled into a tube blank, 1.268 cm (0.499 in.) O.D. x 0.873 cm (0.344 in.) I.D., with the wall consisting of nearly equal thicknesses of Type 304 stainless steel and vanadium. The drilling operation seemed to have no observable effect upon the bond between the steel and vanadium.

After the drilling, the tube was ironed on a nondeformable mandrel to 1.029 cm (0.405 in.) O.D. x 0.850 cm (0.335 in.) I.D. (approximately 60% reduction). The tube will be fabricated further after evaluation by nondestructive testing.

c. V-10 w/o Ti (TV-10) and V-20 w/o Ti (TV-20) Alloys. Two billets of TV-10 and TV-20 were canned in Type 304 stainless steel and extruded at 1110°C and a 7.7:1 reduction ratio. Radiography of the extruded bars indicated a rippled O.D. of the core material, indicating that the V-alloy material was significantly stiffer than the stainless steel at the extrusion temperature. Based upon extrusion pressures, both V alloys are stronger than Type 304 stainless steel, with TV-20 being the stronger of the two. The extruded bars will be warm straightened and dejacketed to provide samples for hot tensile, compatibility, and corrosion tests.

3. Corrosion of Fuel-cladding Materials

A group of five niobium-base alloys have been tested in flowing (0.15 m/sec) sodium for 158 hr at 650°C. Zirconium turnings were used to getter oxygen during exposure of the test samples. The oxygen concentration of the gettered sodium was about 120 ppm.

The alloys tested (composition given in weight percent) are as follows: (1) Nb-5 Mo; (2) Nb-10 Ti-5 Zr; (3) Nb-18 Ti-4 V; (4) Nb-3 Mo-9 Ti; (5) Nb-10 W-2.5 Zr.

Recrystallized and stress-relieved samples of the above alloys were tested to determine the effect of metallurgical condition on the rate of attack. A detailed metallographic analysis is in progress, but qualitatively it was found that the recrystallized samples were more severely attacked and suffered intergranular penetration to a greater extent.

4. Nondestructive Tests of Experimental Fast Reactor Components

a. Tubing. Conventional pulse-echo shear wave techniques were applied in the testing of one tube, Nb-10 w/o Mo-1 w/o Zr-0.07 w/o C, nominally of 0.80-cm O.D. and 0.04-cm wall, for longitudinal and transverse defects. The I.D. reference notches were 0.01 cm deep x 1.27 cm long. The test was performed with a focused 5-Mc transducer. No defects were detected.

Nondestructive tests on tubing being used or produced for general fast reactor fuel development and the results of these tests are summarized in Table VIII.

Table VIII. Data and Results for Tests on Stainless Steel Tubing and Hastelloy-X Tubing

Material	Dimensions	Quantity, m	Test Method	Results
Type 304 stainless steel	4.22 mm I.D. x 0.228 mm wall	32	Eddy current	3 defects $\geq 10\%$
Same as above			X-radiography	No defects noted
Type 304 stainless steel	4.22 mm I.D. x 0.381 mm wall	41	X-radiography	No defects noted
Type 304 stainless steel	4.22 mm I.D. x 0.508 mm wall	34.1	X-radiography	No defects noted
Hastelloy-X	4.22 mm I.D. x 0.254 mm wall	29.3	Eddy current	2 defects $\geq 10\%$

Radiographs were made to determine the amount of tungsten deposited on Hastelloy tubing of 4.22 mm I.D. x 0.38 mm wall. This was done by radiographing the tubes on fine grain film along with known thicknesses of tungsten foils and a Hastelloy tube of the same dimensions as that of the pre-coated tubing. After film processing, density measurements were taken at various points on the coated tubes and compared with that of density measurements taken of the tungsten foils and Hastelloy tube. The results of the measurements showed that the tungsten deposit varied from 0.127 mm to 0.165 mm on one set of tubes, and from 0.122 mm to 0.145 mm on the other.

b. Vanadium-20 w/o Ti Machined Casting. This casting, nominally of 3.49-cm O.D., was ultrasonically inspected at 1 Mc for transverse and longitudinal defects. The reference notches were 0.04 cm deep x 1.27 cm long. Three transverse cracks, located about 10.80 cm from the numbered end, were revealed by a shear-wave technique. One crack produced a defect signal amplitude equal to that obtained from the transverse reference notch. The other two cracks were not as deep or long but still were readily detected by the inspection system.

C. EBR-II

1. Reactor Plant

The two safety rods and twelve control rods were loaded into the reactor and the time-displacement characteristics of the safety rods were determined. The acceleration was determined to be 0.75 g.

A time-displacement curve during scram was obtained for each of the twelve control rods scrambled individually and also for one rod when twelve were scrambled together. The maximum acceleration for each rod was set at between 1.4 and 1.5 g, with an assisting air pressure of 30 psig.

The control rod lifting platform was adjusted, as were the gripper jaw mechanisms for the control rod drives.

The fuel gripper was placed back in operation after replacement of some temporary wiring and some worn parts in the control assembly.

Adjustments were made to the Fuel Unloading Machine to improve the reliability of the "jaws loaded" indication, as well as the alignment with the transfer arm at the "pickup" position.

A special control rod with natural uranium elements was preheated in the Fuel Unloading Machine and heated in the fuel transfer port to determine the effectiveness of heating this type of subassembly. The subassembly

was marked with heat-sensitive "paint." Nine capsules containing 50/50 solder (melting point of approximately 428°F) were placed in "fuel elements" in the subassembly.

Two tests were made, one with a heating time of 45 min and one with a heating time of 20 min. The rod was first preheated until the outlet argon gas temperature of the Fuel Unloading Machine was 205°F. The argon temperature at the inlet to the fuel-transfer port averaged about 665°F.

In the 45-min test, surface temperatures of the element were between 450 and 500°F, and all solder pieces were completely melted. In the 20-min test, surface temperatures of the element were slightly under 450°F, and all but two of the solder pieces had melted completely. Gas flow rates during heating were about 45 scfm.

Twenty-nine core-type subassemblies were loaded into the Fuel Unloading Machine and placed in the storage basket in preparation for wet critical. A checkout procedure demonstrated that the fuel-handling system can be operated without causing spurious scrams, and also that by introducing selected scram signals during fuel handling, the safety rods will indeed scram and the fuel-handling operations will stop. With the fuel-handling shutdown string energized, the following operations were accomplished:

1. Three core-type natural uranium or steel dummy subassemblies were transferred from the core to the storage basket and back to the core.
2. The three sources in the beryllium thimbles were removed and re-inserted in these thimbles. Two of the sources were transferred to storage thimbles. The third source was simply removed from the beryllium thimbles and held by the fuel gripper.

During the above operations, only one spurious scram occurred. It occurred during plug rotation in conjunction with the source manipulations. The trip was due to a noise signal in a period channel.

As part of the checkout, five selected scram signals were interjected to demonstrate shutdown capability by scrambling the safety rods and stopping fuel-handling operations. These signals were: low flux; period and high flux in Channels 1, 2, and 3; blanket argon pressure high; manual scram. All signals scrambled the safety rods and stopped fuel-handling operations.

A background count rate was made for Channels 1, 2, 3, G, and H with the three sources inserted, removed, and replaced in the beryllium thimbles.

The building airlock doors were placed in operation after being successfully leak-tested.

On October 30 the wet critical experiments were started by loading fuel elements to the reactor grid. At month's end 16 elements had been loaded into the reactor.

2. Sodium Boiler Plant

At the beginning of the month, heatup of the system with a simulated failure of the shell heater was in progress. Results show that if a shell heater fails at least 4 hr elapse before dangerous temperature differentials are reached between the shell and water tubes.

When the system was filled with sodium and heatup started, the heat losses from the piping and blowdown proved to be so large that the temperature could not be raised above 400°F when water circulation was maintained through the boiler. The steam system in the Sodium Boiler Plant was then isolated from the Power Plant and yard piping, and the heatup continued. The system temperature increased from 440°F to 490°F in $8\frac{1}{2}$ hr. In this period, the plugging temperature increased from 325°F to saturation (490°F). The system was then dumped and the steam system cooled to 350°F. The sodium in the storage tank was heated to 650°F, cold-trapped to a plugging temperature of less than 300°F, and then cooled to 360°F. During this time, a sintered stainless steel filter was installed in the line leading to the plugging indicator to remove particulate material which was causing malfunction of the plugging meter and contaminating bulk-sodium samples.

The system was then filled with sodium, the steam system isolated from the yard, and heatup with circulation started. When 420°F was attained, the temperature was held constant while measurements of pipe deflection were made. After these measurements were completed, heatup continued until a temperature of 500°F was reached. At this time, the temporary cold trap in the yard piping plugged completely, so the system was dumped. At the time of dumping, the plugging temperature was 440°F.

Throughout this last system heatup, operational difficulties were experienced with the cold trap. A steadily increasing pressure drop across the crystallizer tank was observed. At first, this could be alleviated by a temporary reversal of flow through the cold trap. However, as time went on, this expedient became less and less effective. When the system was

dumped, practically all of the sodium was dumped through the cold trap in a reverse direction in an attempt to wash out, or disturb, the plugging material. After the dump, however, the pressure drop was as high as before. By running the cold trap isothermally, it was shown that the plugging material was not sodium oxide.

Since some flow was possible, the sodium in the storage tank was cold trapped until complete plugging occurred. By using the expedient of temporarily reversing flow through the cold trap, the large pressure drop in the crystallizer tank began to lessen; at present, the pressure drop is normal. The plugging temperature has been reduced to about 250°F.

Previously, trouble had been experienced with particulate matter in the plugging loop and sodium samples. To determine the cause of the high pressure drop in the cold trap, the sintered stainless steel filter installed in the line to the plugging loop was removed. Inspection showed the filter contained considerable quantities of finely divided magnetic material, possibly magnetic iron oxide and/or iron. Next, a magnet was lowered to the bottom of the sodium-storage tank. When retrieved, a quantity of attached magnetic material was found.

It is postulated that as the system temperature is increased the piping increases in diameter due to thermal expansion. When this happens, the brittle mill scale (magnetite) on the inner surface of the ferritic pipe breaks off and is carried in suspension in the flowing sodium. Based on the results of chemical cleaning of the steam system, it is estimated that there is 450 lb of mill scale (magnetite) in the secondary sodium system.

At present, the temperature of the sodium in the storage tank is being raised to determine the temperature at which the sodium will start to reduce the magnetite.

During this period, many analyses of the argon blanket gas were made using a gas chromatograph. A check analysis run on a mass spectrometer indicated that helium gas was also present. Since the chromatograph cannot distinguish between helium and hydrogen and since the solubility of helium in sodium is not known, the previous results are invalid. Work continues toward removing these difficulties.

3. Power Plant

The primary function of the Power Plant during October was to support operations in the Sodium Boiler Plant by supplying feedwater at desired flow rates, temperatures, and pressures.

Early in the month shell heater tests of the steam generator required stepwise increases in feedwater temperature. The initial blowdown rate of 30,000 lb/hr had to be reduced because of excessive heat loss and was eventually stopped completely. Steam-drum level was than satisfactorily regulated by the level controller which loaded and unloaded the startup feedwater pump as necessary.

After sodium was drained from the secondary system, the system was stabilized at 350°F, and circulation of feedwater was resumed with the steam generator flooded. Sodium again was pumped into the secondary system. A water level slightly above the center line was established in the steam drum by draining boiler water. Superheaters and steam main were drained at this time. Stop valves in the feedwater and blowdown lines were then closed to isolate the steam generator for heat-up. Level in the steam drum was maintained by intermittent addition of feedwater as required by losses.

This condition was maintained for 7 days after which the steam generator was cooled to 360°F and flooded. The cool-down was then completed with circulation of the boiler water by the startup feedwater pump. Following this, the steam generator was put in wet layup.

During the week of October 9, the chemical treatment of the boiler water was changed from the sulfite-phosphate system, used for oxygen-scavenging control during steam generator wet layup and low-temperature operation, to the hydrazine-morpholine system, to be used for operations at temperatures around 580°F. Injection rates of the chemicals were increased just prior to isolation to provide ample treatment during static heat-up of the steam generator. With the steam generator in wet layup, the water is treated with morpholine, hydrazine, and sodium sulfite.

4. Fuel Cycle Facility

The trolley for one of the 5-ton cranes in the Argon Cell, which had become inoperable during September, was removed for examination and repair. A short circuit in one of the mineral-insulated cable terminals was found and the cable replaced. The crane is again in operation.

The lifting chain for the lid of the transfer lock cover in the Argon Cell has twice dropped from its drive sprocket. Although the cause is not yet known, it may be associated with an inadequate idler guide. The chain was replaced by use of the General Mills Model-300 arm attachment for the electromechanical manipulator.

Continued deterioration in the activity of the catalytic bed for the Argon Cell purification system is indicated by slow removal of oxygen. Although analytical instrumentation is temporarily not working well, there

appear to be several hundred ppm oxygen in the cell gas, with about 15 ppm water and an apparent value of about 8 percent nitrogen. (The nitrogen instrument is also responsive to the presence of hydrogen and helium) Tests are being conducted to evaluate the activity of the catalyst.

Measurements of piston ring wear in the dry argon compressors indicated excessive decreases in thickness (to only 7-20 mils clearance), and fluffy carbon deposits were noted in the cylinders. The rings, of carbon-filled Teflon, will be replaced.

The following fuel fabrication equipment is now operational: Charge Table; Pallet Load and Unload Station; Injection Casting Furnace; Fuel-rod Assembly Machine; Settling Machine; Bonding Machine.

The dismantler and its supporting equipment, designed to carry out the remote disassembly of the core subassemblies, have been installed in the Air Cell (see Figure 4). All cutting operations used in the process are chipless and the cooling system utilizes suction flow to reduce the spread of particulate contamination. Radiation-resistant materials have been used for most in-cell components of this machine; some of the exceptions are the electric motors, lubricants, and the static seal on the air cylinders. The units incorporating these materials (allowable exposure about 10^9 r) are easily removable to facilitate remote repair.

Figure 4. EBR-II Core Disassembly Dismantler as Installed in the Air Cell of the Fuel Cycle Facility. A fuel subassembly is shown being rotated to the horizontal position.

The machine, as now installed, is equipped to handle fuel subassemblies by use of the Model 8 manipulators equipped with special tongs to remove the fuel elements from the support grid (see Figure 5). Because of the time and potential human error involved in identifying each fuel element, along with the possible need of exerting considerable force to remove the fuel elements, a specific-purpose Fuel Element Removal Machine has been designed and fabricated. This unit is undergoing tests at this time and will supplement the hand operation when its reliability has been proven.

Figure 5. An Attachment to the Tong of the Master-slave Manipulator Is Being Used to Remove a Fuel Element from the Grid Support Rail of a Fuel Subassembly.

The disassembly of six other subassembly configurations (blanket, control, safety and experimental) is achieved with the use of attachments which are being fabricated.

The dismantler cooling system, consisting of the suction blowers and high-efficiency filter bank located in the subcell area, together with the connecting line, in-cell filter and headers, suction-head sensors and flow monitors, is used to draw air through the subassembly axially or transversely during all stages of the dismantling process. An operational test after installation indicates the design requirements of 3 kW of heat removal, with a maximum fuel element temperature of 600°F, have been met with a 30 percent overcapacity safety factor. It will be necessary to further muffle and isolate the cooling system to reduce noise transmitted through the suspect air-exhaust system which extends to several areas of the building. The noise is estimated to be approximately 70 to 80 db at a 90-cps fundamental pitch.

All important functions of the machine are interlocked with the position of the cooling flow valves on the dismantler to prevent accidental

loss of cooling air flow during operations. The operation of both the valve function switch and a flow-actuated switch is necessary to clear critical interlocks.

The dismantler was installed remotely in about 10 days by operating personnel using the installation manual as a guide.

A preliminary run-through of the operations for the dismantling of one fuel subassembly required 5.5 hr. This involved the use of the master-slave manipulators for removing and loading the fuel elements. Subassembly and magazine transfers are estimated to require an additional 1.5 hr. Operation and repair manuals, illustrated with photographs have been prepared.

The spiral decanning machine removes the stainless steel hardware from the fuel pin and breaks the pin into short lengths. Two complete spiral decanning machines have been delivered to the Fuel Cycle Facility. One of these machines has been installed in the Argon Cell at window 15 and is presently being used to decan sodium-bonded elements (see Figure 6). The second unit is used for out-of-cell testing and provides a complete set of replacement parts.

Figure 6. EBR-II Fuel-decanning Machine

All of the decanning operations have been automated by the use of electric motors and electrically controlled pneumatic actuators. The operations are shown schematically in Figure 7. The stainless steel jacket is shown being sheared from the element by a single point tool in Figure 8.

Figure 7. EBR-II Fuel-decanning Operations

Figure 8. Detailed View of the Rotary Shearing Operation Used on the Spiral Decanner. The stainless steel tubing is sheared into a spiral strip by the tool and the cutting roll.

About 8 hr were required to assemble the machine after the parts had been transferred into the cell. Movies and photographs have been taken of the installation and incorporated into an operational manual.

The portable control console containing all relays, switches, and timers used in the circuitry is located on the operating floor at window 15. The solenoid valve bank is installed in the subcell area. Fuel elements may be decanned individually with the operator initiating each of the ten machine functions, or the operations may be grouped so that several fuel elements are in the decanner at once and only three steps are needed per fuel element. A repeat mode is also included which allows the operator to start one element through the machine and then the entire magazine of 25 elements is automatically processed.

A number of performance tests were carried out on the decanner before it was shipped to Idaho: (1) the machine decanned elements with broken fuel rods (0.125- to 1.250-in. pieces); (2) maximum steel contamination of 24 ppm by weight could occur if all of the dust produced entered the fuel container; and (3) a recent modification to the cutting tool and actuating mechanism enabled the machine to decan elements having the tubing flared over on the ends.

After installation in the argon cell, about 150 elements have been decanned, of which 40 were sodium bonded. Some difficulty has been encountered with the fuel-scrap separator tube, scrap cutter, and fuel-pin tray. All of the problems appear to be due to the effect of cold sodium adhering to cold metal surfaces in the extremely dry atmosphere. The effects of sodium on an earlier version of the decanning machine were not as severe, since the moisture content was usually 30-100 ppm. Installation of a sealed Model A master-slave manipulator has greatly facilitated overcoming operating problems. Removal of the decanner drive unit revealed considerable wear, which was corrected by fitting, shimming, and tool replacement, following which a group of 20 elements was decanned satisfactorily. Testing is continuing on use of heat to alleviate problems of sodium accumulation. The scrap from decanning the first group of 91 elements was found to contain 27 g of uranium in the form of small chips which appeared to have been the result of chopper action.

In further operations with the injection-casting furnace, a small group of castings of acceptable length was obtained. A balance table has been fabricated and is ready to be located in the cell with the 20-kg balance. The pin-processing machine has been used successfully to remove glass molds from castings, although short castings are difficult to handle. Studies of separating crushed Vycor glass from scrap uranium by air flotation are being made. Both can welders have been modified, and one was installed in the Argon Cell; successful welds have been made

remotely. Shorting of high-frequency feedthroughs for the welders has led to a temporary suspension of operations. A leak-testing method has been devised for routine use on new fuel element cans, and can volumes are being measured for evaluation of sodium loading.

The bond-and-level-inspection machine has been installed in the air cell and operated mechanically. Good temperature control has been achieved.

Two of the shielding doors for the analytical laboratory cells have been modified by replacing defective lifting screws and nuts with ball nut screws, and rework of the remaining doors is underway. Standardization of the mass spectrometric method for burnup determination in terms of lanthanum-139 is proceeding. The gamma spectrometer has been calibrated for the determination of molybdenum-99 in fission foils.

Leak tests of the Argonne-fabricated interbuilding fuel-transfer coffin (see Progress Report for September 1963, ANL-6784, p. 21) have indicated that the leakage rate will be less than one cu in./hr at one-psi air pressure in the coffin. Performance tests under plant-operating conditions are in progress to determine the useful operating period between charges of the battery which provides power for the motor-blower assembly for the recirculation of cooling gas in the coffin.

Layout drawings of a blower system and associated controls for cooling Air Cell storage tubes and the spent or refabricated fuel subassemblies which the tubes will contain have been completed (see Progress Report for September 1963, ANL-6784, p. 22). The system will consist of two lobe-type blowers, each of which can deliver approximately 135 cfm air at about 2 psig. Only one blower will be in continuous operation. The second blower is provided for standby operation and will automatically be placed in operation should the other motor or its blower fail. The blower system and controls will be built and tested at Argonne before being sent to Idaho. Equipment items have been ordered.

Construction drawings of the skull-oxidation furnace for use in Idaho have been completed. The furnace and associated control system and panel board will be fabricated and tested at Argonne before being shipped to Idaho.

5. Process Development

a. Skull Reclamation Process. The skull reclamation process will be used for the recovery and purification of the fissionable material that remains in the melt-refining crucible after a melt-refining operation. The steps include: noble metal extraction, uranium reduction, precipitation of a uranium-zinc intermetallic compound, intermetallic decomposition,

uranium dissolution, and (after removal from the primary vessel to a separate vessel) retorting of the uranium product solution to concentrate the uranium. Except for the last step, each step is followed by a phase separation, in which a molten metal or flux phase containing impurities is separated from a uranium-bearing phase by pressure siphoning through a heated transfer line. All steps of the skull-reclamation process (except the final step of isolating uranium metal by evaporation of solvent metals) are presently being conducted in a single 20-liter tungsten crucible.

In one step of the process, a zinc-uranium intermetallic compound is precipitated from a uranium-zinc-magnesium solution by cooling the solution without agitation from 800°C to about 500°C. The supernatant solution is then removed and discarded. In previous runs, low efficiencies of removal of the supernatant (60 to 70% instead of a desired 80 or 90%) were experienced during this process step. Four runs, which duplicated the intermetallic precipitation step only, have recently been performed in an attempt to solve this problem. In each of the runs, the uranium-zinc-magnesium mixture was agitated at 40 rpm as it was cooled from 800°C to 600°C. The melt was then cooled from 600°C to 500°C without agitation. Supernatant removals in these runs ranged from 84 to 89%.

b. Blanket Processing Studies. The engineering-scale distillation unit for studying the retorting of magnesium-zinc-plutonium supernatant solution generated near the end of the EBR-II blanket process has been modified. The position of the heating coil on the $8\frac{3}{4}$ -in.-diameter, 8-in.-high still pot was varied in order to find the best position for refluxing of distilled metal and decreasing deposition of solute on the still pot wall. Three runs were performed with 8- to 10-kg charges of 50 w/o magnesium-zinc containing about 0.5% tin as a stand-in for plutonium. The best refluxing occurred in a run in which the induction heating coil was positioned to provide heating of the bottom 3 in. of the still pot wall. At the end of this run, the deposit of tin on the still pot wall was only 0.2% of the tin charged as compared with a deposit of 29 and 32% of the tin charged in two runs in which the lower 5 in. of the still pot wall was heated.

c. Sodium Coolant Chemistry. Methods for the purification of sodium are being developed in preparation for study of the reactions of liquid sodium. A small all-metal apparatus has been built to prepare high-purity sodium. The unit consists of: a cold trapping vessel where impure sodium is heated just above its melting point, degassed, and filtered through stainless steel wool; a sodium distillation unit; and a vacuum chamber for holding a crucible while it is being filled with purified sodium.

Experiments are in progress to produce sodium of low carbon content by other methods. The methods being studied include: (1) "gettering" of the carbon and oxygen in sodium by contact with zirconium chips at elevated temperatures and (2) thermal decomposition of sodium azide.

Experiments are being performed in an effort to correlate three methods of determination of oxide content of sodium, namely, through use of the plugging meter, the Blake meter, and distillation. The sodium oxide analysis made with the plugging meter on the same sample of sodium varies with the temperature of the main flowing circuit. These analyses did not agree with those obtained from the distillation method.

6. Training

A group for initial reactor operations, comprised of the reactor manager, six shift supervisors, four foremen, and a reactor operator, completed the training for wet critical. They are now authorized as qualified reactor operators for wet critical operations in accordance with ANL procedures.

Six 8-hr shift periods were used expressly for control room practice to complete training for wet critical functions. The initial group of qualified reactor operators were participants in the dry critical experiments.

A second group of trainees have met all requirements for wet critical qualification except actual nuclear operation of EBR-II.

Wet critical training for additional shift reactor operator trainees has progressed intermittently because of the priority on completion of training for the first two groups mentioned above.

D. FARET

1. Design Studies

The FARET Title-I Design Report has been completed and transmitted to the Commission for approval to proceed with Title II. In the meantime, studies are being made of the following:

- a. a review of the Title-I engineering and construction schedule with reference to engineering manpower leveling and work packaging;
- b. a review of the procedural requirements for use of a PERT-type schedule to monitor progress;
- c. construction of a rather simplified working model of the FARET cell to serve as a visual aid during the design of in-cell equipment (the model will be used to resolve viewing problems, equipment arrangement location, location of services, and penetration considerations);

- d. reference drawings of the cell to provide a means for coordination of cell and cell-equipment design parameters;
- e. the continued review of the preliminary design of the reactor vessel.

2. Safety Analysis

The analogue studies described in ANL-6784 (Progress Report for September 1963, p. 26) have been completed by Bechtel.

The effect of the radiant heat transfer mechanism upon cell pressure during a sodium-air incident within the cell was evaluated with use of an IBM-704 code (1922/RE). Radiant heat transfer coefficients for conditions such as may exist are not known; however, coefficients have been assumed which are considered as upper and lower bounds.

The peak temperature, and hence pressure, in the cell were determined with use of the following assumptions:

- a. There is an instantaneous reaction at time = 0.
- b. Sodium combines stoichiometrically with all oxygen and water vapor in the enclosure.
- c. The peak temperature is the same for all reactants and is achieved at zero time.
- d. Specific heats of reactants are taken from ANL-5750.¹

The temperatures and pressure as a function of time were then calculated on the basis of the following assumptions:

- a. Heat loss is to a liner by convection and/or radiation (gas radiation constants are not calculated).
- b. There are no heat losses from the gas-liner system.
- c. The liner and gas are at uniform temperatures at any time.
- d. There is no gas leakage from the enclosure.
- e. The perfect gas law is valid.

¹A. Glassner, A Survey of the Free Energies of Formation of the Fluorides, Chlorides and Oxides of the Elements to 2500°K, ANL-5750 (1957).

Three conditions of heat transfer were studied. Each was based upon the physical constants of the FARET cell: an initial gas temperature of 100°F, an initial sodium temperature of 300°F, a gas moisture content of 0.0108 lb/lb, and a stoichiometric reaction of sodium and air. The stoichiometric reaction was used because the present computer code can handle it most conveniently. The credibility and magnitude of sodium-air reactions are still under study for FARET. The three cases considered were:

- a. convection cooling only;
- b. convection cooling plus "pessimistic" loss of heat by radiation;
- c. convection cooling plus "optimistic" loss of heat by radiation.

The results are shown in Figure 9. The initial conditions in each of the three cases are peak temperature and pressure of 6422°R (5962°F) and 111.2 psia, respectively. The final equilibrium temperatures are 647°R (187°F) and 11.2 psia, respectively. The equilibrium pressure is below atmospheric (12.2 psia) because all the oxygen is consumed in the reaction. The significant contribution of radiant heat transfer is quite apparent.

Figure 9. Pressure-time Profile for Stoichiometric Na-air Reactor with 0.0108 lb H₂O/lb Air

3. In-core Instrumentation

Tungsten-rhenium alloy thermocouples are the reference design sensors for the measurement of fuel pin temperatures in the FARET reference design. These thermocouples must be terminated at the proposed in-vessel connector in base metal pins because the refractory metals have

been difficult to fabricate into suitable contacts. The splice between the thermocouple wire and the pins or extension lead wire at the in-vessel connector will operate at temperatures in the range between 250 and 650°C. It appeared from the literature that a 18-8 stainless steel/alumel combination might serve as a suitable extension lead wire. To verify this, three sample stainless steel (Type 308 ELC weld rod)/alumel thermocouples assemblies were fabricated. These were calibrated in an oven up to about 650°C. The results are shown in Table IX. These results verify the data found in the literature and give some assurance that this lead wire combination closely matches the thermoelectric properties of the sensor.

Table IX. Type 308 ELC Stainless Steel/Alumel Lead Wire Characteristics

(Reference Junction at 0°C)

Temp, °C	Output, mV					Date
	W-3% Re	308 ELC Stainless Steel			Average	
	W-25% Re	Alumel				
151	1.86	2.49	2.49	2.50	2.49	10/4/63
198	2.57	3.17	3.18	3.20	3.18	
253	3.48	3.96	3.97	3.99	3.97	
300	4.29	4.63	4.65	4.68	4.65	
360	5.37	5.59	5.61	5.64	5.61	
415	6.42	6.47	6.49	6.53	6.50	
470	7.50	7.44	7.46	7.51	7.47	
529	8.67	8.53	8.56	8.61	8.57	
580	9.69	9.54	9.56	9.62	9.57	
643	10.96	10.84	10.87	10.94	10.88	
152	1.87	2.53	2.54	2.54	2.54	10/9 and 10/10/63
202	2.63	3.22	3.23	3.25	3.23	
259	3.58	4.06	4.06	4.09	4.07	
311	4.48	4.81	4.83	4.85	4.83	
360	5.37	5.59	5.60	5.63	5.61	
420	6.52	6.53	6.56	6.58	6.56	
476	7.62	7.54	7.56	7.60	7.57	
528	8.65	8.48	8.50	8.54	8.51	
588	8.85	9.65	9.67	9.73	9.68	
640	10.90	10.67	10.78	10.83	10.76	

Both stainless steel and alumel are standard pin materials used in commercial connectors. In order to evaluate completely the adequacy of this extension lead wire, it will be necessary to operate a sample connector and thermocouple under simulated FARET operating conditions. Other lead-wire combinations which might better match the tungsten-rhenium alloys over a wider temperature range will be sought and tested.

4. Fuel Development

As noted in ANL-6784 (Progress Report for September, 1963, p. 27), use of "slip-fit" fuel pellets for Doppler measurements is being considered because the pellets which are insulated from the cladding by a gas gap may crack. Slip-fit pellets require higher heat fluxes than gas-gap pellets to obtain a desired fuel temperature change. Therefore, in order to test slip-fit pellets "in-pile," it is necessary to provide larger capability for heat removal than for gas-gap pellets. An in-pile facility which had been designed during the past few months for testing gas-gap pellets was found to be inadequate for this purpose. The facility is being redesigned to accommodate pellets of both high and low heat-flux output. Figure 10 illustrates the current design intended for use in CP-5.

Figure 10. FARET Fuel-irradiation Arrangement

The fuel element capsule, consisting of pellet and clad, is inserted in a jacket which forms a 0.006-in. radial gap. This gap is filled with helium gas at a pressure varying from atmospheric to $10\ \mu$ in order to change the clad temperature as desired. The annulus formed by the thimble and the jacket is filled with D_2O so that heat from the pellets passes first through the clad then successively through the helium gas gap, jacket, D_2O annulus, and thimble wall to the CP-5 reactor D_2O as a heat sink. The helium gas-gap method will be used for the low heat-flux output. For high heat-flux pellets, a 0.012-in. gap, replacing the 0.006-in. gap between clad and jacket, is filled with sodium as a thermal bond, and the clad temperature is varied by changing the helium pressure on the annulus D_2O . (With high heat-flux pellets the D_2O boils, and changes in saturation pressure, accordingly, change the jacket-surface temperature.)

Capsule power is determined by two thermocouples positioned in the annulus D_2O , one at the level of the lower end of the fuel and one 2 in. below. Laboratory tests indicate that the difference between the temperature at these two positions varies linearly with capsule power (about $0.5^\circ C/cm/W$) up to the point where the upper temperature approaches the saturation temperature of the D_2O . The thermocouples will be calibrated in-pile with an electrical heater.

When the low heat-flux pellets are enriched with plutonium in the same manner as the reactor fuel, it will be necessary to reduce the reactor flux by about a factor of 10. This will be accomplished by installing a perforated cadmium sleeve, in an aluminum jacket, outside the thimble. Tests of such a neutron filter are now under preparation for the CP-5 reactor. The effectiveness of the filter will be determined by measuring the fission rates in a uranium foil with and without the filter.

Other fuel concepts are also being examined. Two concepts, both of which use a straight ceramic pellet, have been proposed. In one concept the pellet is supported by a fluted can; in the second, the pellet is supported by a metal helix.

Work also continues on the warm pressing of UO_2 grains bonded with glass to produce specimens having densities in excess of 90% of the theoretical density of UO_2 .

The sintered UC pellets for heat-flux bench tests are ready for assembly. Some difficulty was encountered in drilling the holes in these specimens, and a number of the samples cracked.

5. Doppler Measurements in FARET

An experimental program to measure the Doppler effect in the FARET reactor facility is being developed for an oxide-fueled uranium blanket reference reactor.

Several methods are being explored to confine a significant temperature change in the fuel elements of the test zone. One of these methods, requiring a minimum of developmental effort, is the varying-power-level experiment without a fuel-clad gas gap.

In this method fuel pellets are slip-fitted inside the clad. The fuel temperature change is then dependent on the power density in the rod, the conductivity, and the radius of the fuel pin. The changes of coolant and clad temperatures in the test zone are kept small by varying the coolant flow rate in direct proportion to the change in the power level. The temperatures in the buffer and driver region are maintained essentially constant by this increased coolant flow.

The poor conductivity of the uranium oxide allows a significant temperature increase in the fuel pin. An oxide reference core having a U^{238} to Pu^{239} atom ratio of 7 was taken as being typical of large (2000-liter) systems. The Doppler coefficient, or rather the Doppler component of the fuel temperature coefficient, for this system was computed to be $-7.3 \times 10^{-6} \Delta k_{\text{eff}} / \Delta T^\circ\text{C}$ at 750°K . This is considered to be of significant magnitude to generate a measurable reactivity effect in the FARET zoned system.

Of the many multiregion configurations studied, the system that appears to satisfy most of the conditions, is one where the outer radii of the test zone, buffer region, and driver section are 7.4 in. (18.8 cm), 13.7 in. (34.8 cm), and 15.8 in. (40.1 cm), respectively. The buffer region contains sodium-cooled, depleted UO_2 pellets, with the volume fraction of the oxide/sodium/stainless steel structure kept the same as in the test zone. The remaining region of the FARET system contains sodium-cooled stainless steel subassemblies.

The neutron spectrum matching was kept to within 10% of that in the corresponding region of the reference core.

A criterion used was that a reactivity change arising from a change in the fuel temperature of the test zone be within some acceptable error of the reactivity change in the test zone of the reference core. This assures one that the importance spectrum as well as the neutron spectrum is reasonably reproduced.

Another condition imposed on the FARET experiment is that the diameter of the fuel rod used in the test zone be as close to the rod size that would be used in the actual power reactor but still allow the desired fuel temperature change. There is no limit to the fuel temperature rise that can be obtained in the varying-power-level experiment, since one can increase the size of the rod. However, in order to reduce possible heterogeneity effects on the Doppler coefficient, the size of the rod should be maintained as small as possible.

To optimize the signal in this experiment, a fuel temperature increase of greater than 500°C is desired. The average power density in the test zone of this system is $0.061 \text{ MW}/\ell$, which will allow the desired temperature rise for a $\frac{1}{2}$ -in. (1.27-cm)-diameter pin. More compact zoned systems are being studied in an effort to obtain a temperature change greater than 500°C with fuel pin diameters less than $\frac{1}{2}$ -in. (1.27 cm).

The temperature dependence of the averaged Doppler coefficient was maintained to within 20% of that expected in the reference core. This reproducibility was maintained over a temperature range from 300°K to 1500°K and estimated to hold up to 2500°K .

The problem of isolating an interpretable Doppler effect in a reactivity measurement depends on the contributions to the signal by effects due to the fuel expansion in the test zone, and the temperature changes of coolant and structure in the whole system. Therefore, an integral part of this experiment is a rigorous error analysis.

Averaged sodium- and fuel-expansion coefficients were determined for the entire zoned system, including the reflector. The errors associated with these coefficients were based on variations observed in the calculations of many-zoned systems having a variety of spectra, including systems studied with slightly different sets of cross sections. Average temperature changes which can be expected in varying the power level by a factor of ten were also computed. The errors associated with these temperature changes are estimates based on thermocouple experience and at present are judged to be reasonable, at least in the 1500°C temperature range.

The expected Doppler reactivity change for a conservative fuel-temperature increase of 500°C is $-410 \times 10^{-6} (\Delta k/k)/^{\circ}\text{C}$. From the error analysis, the standard deviation was found to be about 12%, assuming that a series of determinations to give the expected experimental value of $-642 \times 10^{-6} (\Delta k/k)/^{\circ}\text{C}$ has an error of 10%. The Doppler coefficient can then be determined to within 20%. Even if the errors associated with the extraneous signals were relaxed, the Doppler component would still be observable in the total signal.

To establish more firmly the extraneous contributions and associated errors, a consistency check can be made by comparing measured reactivity values with the expected values at various temperatures.

Studies will be continued on the design of a zoned system where a temperature change greater than 500°C can be obtained with fuel pin diameters less than $\frac{1}{2}$ in. (1.27 cm).

A system presently being considered is one where the outer radii of the test zone, buffer region, and driver section are 5.3 in. (13.5 cm), 11.6 in. (29.5 cm), and 13.7 in. (34.8 cm), respectively. The power density of this system is 0.074 MW/l.

III. GENERAL REACTOR TECHNOLOGY

A. Applied Nuclear Physics

1. Neutron Scattering by Time of Flight

a. Elastic Scattering. A study of differential elastic scattering has now been extended to include approximately 60% of the periodic table. Scattered neutron resolutions are typically in the range 10-30 keV with

selected measurements at resolutions of less than 1 keV. A typical result in the region of heavy nuclei is the elastic scattering from the standard material, gold, shown in Figure 11. The cross section is expressed in the form

$$\frac{d\sigma(\text{el})}{d\Omega} = \frac{\sigma(\text{el})}{4\pi} \left(1 + \sum_i P_i W_i \right),$$

where the P_i are Legendre polynomials and W_i coefficients determined from the experiment. The inelastic contribution is completely resolved from the elastic component described here.

The available experimental information can be used to determine the elastic transport cross section for a wide range of incident neutron energies and mass numbers; the result is shown in the three-dimensional plot, Figure 12, of elastic transport cross section as a function of mass and energy. The effect of closed neutron and proton shells is evident in the detailed structure. In the region of spherical nuclei, the general behavior is in qualitative agreement with the predictions of the optical model of the nucleus including the effect of surface absorption. The transport cross section can be utilized directly in the formulation of the multigroup cross-section sets employed in fast reactor calculations.

Figure 11. The Differential Elastic Scattering Cross Section of Gold

Figure 12 The Elastic Transport Cross Section Given as a Function of Incident Neutron Energy and Mass Number

The cross sections described above are average cross sections in the sense that they do not resolve the detailed resonance structure of the target nucleus. More detailed measurements in the region of medium-weight nuclei made over flight paths of up to 25 m show a very detailed resonance structure, even at relatively high energies. An example of this is the total cross section of iron shown in Figure 13, in which a multitude of previously unknown resonances are evident in the range from 500 keV to 1.5 MeV. The experimental method is capable of exceedingly good resolution with rapid data acquisition; the major problem is the interpretation of the results. Such interpretation is now being studied by means of conventional resonance-analysis methods and by examination of the fluctuations in the resonance structure.

b. Inelastic Scattering. The inelastic cross section of gold is typical of that of a heavy odd- A element. Some of the experimentally determined excitation functions are shown in Figure 14. The solid curves in this figure represent the results of theoretical calculations by Moore and Auerbach employing an equivalent spherical optical potential and Hauser-Feshbach theory. The theoretical excitation curve for the first level is in fair agreement with the experimental data, particularly at higher energies. The doublet near 270 keV was not resolved experimentally; thus

the data points represent the sum of the two theoretical curves. At higher energies, the level structure of gold becomes quite complex and is not shown here.

Figure 13. High-resolution Transmission Cross Section of Iron

The large number of inelastic measurements now completed make it possible to establish the nonelastic cross section for a wide range of mass numbers and incident neutron energies. These are compared in Figure 10 with the predictions of the nonlocal optical calculations of Perey and Buck. Here the calculated reaction cross section is indicated by a solid curve. The data points and the dotted-dashed curve represent the nonelastic cross section determined experimentally. Certainly, in those instances where there are a large number of inelastic exit channels available, the nonelastic cross section should approach the theoretical reaction

Figure 14. The Lower-energy Inelastic Excitation Cross Section of Gold

scattered from manganese and zinc were made at eight angles between 20° and 160° , with use of the pulse-shape-discrimination neutron-detector system (see Progress Report for September 1963, ANL-6784, p. 31).

cross section since the compound elastic cross section would be very small. This would seem to be the case as the figure shows the experimental nonelastic cross section to be very close to the theoretical reaction cross section in mass regions where inelastic scattering is pronounced. Since the reaction cross section is the sum of the compound elastic and nonelastic cross sections, the compound contribution can be obtained from theory and experiment. This is indicated by the dashed curve. The general agreement between the theoretically predicted and the measured values given in Figure 15 is very encouraging, perhaps falsely so, as theory utilized a spherical potential to describe nuclei that are known to be deformed.

2. Elastic Neutron Scattering with Pulse-shape Discrimination

Angular distribution measurements of neutrons elastically

Figure 15

A Comparison of the Theoretical Reaction Cross Section of Perey and Buck (solid curve) with the Measured Nonelastic Cross Sections

A neutron energy spread of ~ 20 keV was used and the data were taken from 686 to 1097 keV in increments of 8 keV. The data were corrected for multiple scattering in the scatterer, for detector efficiency, and for the finite solid angle subtended by the detector. The fully processed data were averaged over 60 keV to smooth out some of the fluctuations in the cross section. The results for manganese are given in Figure 16 and for zinc in Figure 17. All angular distributions were separately fitted by the method of least squares to a Legendre polynomial series:

$$\frac{d\sigma(\text{el})}{d\Omega} = \frac{\sigma(\text{el})}{4\pi} \sum_{\ell=0}^4 W_{\ell} P_{\ell}(\cos \theta); \quad W_0 = 1.$$

The solid curves in Figure 16 are the result of a Hauser-Feshbach calculation. It is evident from the figures that appreciable fluctuations remain even when the data are averaged over 60 keV. In the case of manganese, the neutrons inelastically scattered from the 130-keV level in Mn^{55} were not completely rejected. The total elastic scattering cross section $\sigma(\text{el})$ in Figure 16 should be reduced by 10% to 20%, depending on the energy. The precise correction for each experimental measurement has not yet been evaluated.

Figure 16. Elastic Neutron Scattering from Manganese. The neutron energy resolution is ~ 60 keV. The dotted line is sketched in to better indicate the energy-dependent behavior. The solid curves are the result of a Hauser-Feshbach calculation.

Figure 17. Elastic Neutron Scattering from Zinc. The neutron energy resolution is ~ 60 keV. The dotted line is sketched in to better indicate the energy-dependent behavior.

3. High-conversion Critical Experiment

Experiments with very heavily loaded water-moderated-and-reflected cores with 3% enriched UO_2 fuel are in progress. A loading using all the available aluminum-clad fuel (4900 elements averaging 853.3 gm UO_2 per element) loaded into the 1.127-cm pitch triangular lattice did not have sufficient reactivity to reach criticality. However, the removal of one fuel pin from each group of 16, 12, 9, or 6 increased the moderator content sufficiently to achieve criticality under the conditions shown in Table X. The atomic ratios of hydrogen to U^{238} (H:28) were, respectively, 1.31, 1.43, 1.60, and 1.96. It was estimated that several thousand additional fuel elements would be required to achieve criticality with the grid fully loaded. The H:28 ratio for this loading would be 0.986.

Table X. Critical Loadings in 1.127-cm Triangular Lattice

Fuel Elements/Grid Openings	11/12	8/9	5/6
H-to- U^{238} atom ratio	1.43	1.60	1.96
Peripheral fuel pin worth, % $\Delta k/k$ per pin	+0.00275	-	0.00858
In-core pin worth, % $\Delta k/k$ per pin	-0.017	-	-0.0406
Clean critical number of fuel pins	3680 ± 18	2788 ± 15	1862 ± 3
Clean critical loading, kg of U^{235}	83.92 ± 0.40	63.82 ± 0.34	42.62 ± 0.07
Critical radius, cm	37.49 ± 0.09	33.15 ± 0.09	27.96 ± 0.02
Radial reflector savings, cm	10.67 ± 0.40	-	8.86 ± 0.40
Radial buckling, $\text{cm}^{-2} \times 10^4$	24.94 ± 0.42	30.78 ± 0.72	42.66 ± 0.93
Axial reflector savings, cm	22.70 ± 0.50	-	18.03 ± 0.50
Axial buckling, $\text{cm}^{-2} \times 10^4$	4.72 ± 0.03	4.77 ± 0.07	5.04 ± 0.04
Critical buckling, $\text{cm}^{-2} \times 10^4$	29.66 ± 0.42	35.55 ± 0.72	47.70 ± 0.93

In order to eliminate the uncertainty of the water hole effects resulting from the removal of the individual fuel elements in these cores, a new grid of approximately 1.17-cm triangular pitch will be obtained. This will permit the assembly of a uniform core having a H:28 ratio of 1.3. These

data will permit the comparison of experimental data with the results of the calculations published by Craig et al.,² and Bengston and Hellens.³

Initial conversion ratio (ICR) measurements and determination of the epicadmium-to-subcadmium capture ratio in U^{238} (ρ^{28}) are reported in Table XI for cores having a H:28 ratio of 2.9 and 2.3 using stainless steel-clad fuel elements. A sizable correction must be applied due to the gap effect resulting from the insertion of foils and their covers between the fuel pellets. Corrections based upon measurements decrease the ICR and ρ^{28} values by 2% or 3%, but calculated values indicate an 8% correction is required. The uncorrected measured values for ICR and ρ^{28} are significantly below those calculated by Bengston and Hellens (-5% in the 2.3 core and -1% in the 2.9 core).

Table XI. Measurements of ICR and ρ^{28}

Parameter	Value	Number of Determinations
A) 1.24-cm Square Lattice; Stainless Steel Clad; Uniform Core; H:28 = 2.9		
Capture Cadmium Ratio in U^{238}	1.269 ± 0.67	11
ρ^{28}	$3.717 \pm 3.16\%$	11
ICR	$0.514 \pm 2.33\%$	11
α	0.2471	
MCR	$0.641 \pm 2.33\%$	11
B) 1.27-cm Triangular Lattice, Stainless Steel Clad Zone, H:28 = 2.3		
Capture Cadmium Ratio in U^{238}	$1.237 \pm 0.86\%$	8
ρ^{28}	$4.216 \pm 4.47\%$	8
ICR	$0.569 \pm 1.72\%$	12
MCR	$0.716 \pm 1.72\%$	12
α	0.259	

The reported values of the ICR and ρ^{28} are based upon analyses of coincidence counting of foils irradiated in these cores and in standardized thermal columns.

B. Theoretical Nuclear Physics

1. Doppler Effect Studies

Investigations of the effects of interference interactions on the Doppler effect calculation have been made independently by a number of

²D. S. Craig et al., Hazards Summary Report, High Conversion Critical Experiment, ANL-5715-Addendum II (Dec 1960).

³J. Bengston and R. L. Hellens, A Survey of the Beginning of Life Characteristics of Uranium-fueled Water-moderated Reactors, CEND-137 (1961).

workers. A significant reduction in the Doppler broadening of the fissile isotope in the intermediate energy region was observed due to the presence of the fertile isotope in the mixture.

Studies were based on the assumption that the overlapping effect of neighboring resonances would not contribute significantly to the interference interaction. For a U^{235} - U^{238} system, however, this assumption becomes rather doubtful in the low-keV region. A more general expression including the overlapping effect of neighboring resonances has been developed. Numerical calculations can be obtained quite readily by a slight modification of RE-270 code; however, numerical results are not yet available.

2. Analysis of ZPR-VII Data

Calculations of thermal parameters for the Hi-C lattices by the THERMOS multigroup code and various one-group methods have been completed.

Collision probabilities calculated by the 1692/RE code were compared with those calculated by Fukai's exact method and other approximate methods. Collision probabilities were also used to calculate thermal disadvantage factors for tightly packed lattices and the results were compared with those obtained by other methods.

3. Orthonormal Expansion of Neutron Spectra with Foil-activation Measurements

In many cases of interest it is impossible to measure neutron spectra with conventional detectors. For example, in hostile environs one usually has no recourse but to utilize an activation method. This often-exploited technique has certain advantages. Perhaps the most important advantage is the negligible flux perturbation and depression which results from the use of sufficiently thin foils.

It is important to consider the different methods for using activation techniques in developing spectral information. In one method under examination, the activation measurements are used to determine an approximate expansion of the neutron spectrum with an orthonormal set of functions. In this method, the flux of neutrons per unit energy interval, $\phi(\epsilon)$, is assumed to be represented by the form

$$\phi(\epsilon) \approx \sum_{i=1}^n \gamma_i u_i(\epsilon) \quad . \quad (1)$$

The set of functions $\{u_i(\epsilon)\}$ is orthonormal in the customary sense:

$$\int \rho(\epsilon) u_i(\epsilon) u_j(\epsilon) d\epsilon = \delta_{ij} \quad , \quad (2)$$

with $\rho(\epsilon)$ the appropriate weight function and δ_{ij} the Kronecker delta function. The set of coefficients $\{\gamma_i\}$, which occur in the representation of Eq. (1), are determined by foil-activation measurements.

In addition to the errors inherent in the approximation employed in the analysis, there are two other independent sources of error which must also be considered. Since the method relies upon the absolute neutron absorption in each foil material, one must consider any experimental counting error inherent in measuring the activity in the foil. The other source of error is the uncertainty in the cross sections which are employed in the calculations.

A computer program is being developed to investigate this method. The program will permit a quantitative evaluation to be made on the accuracy of the orthonormal expansion technique.

C. Reactor Fuels Development

1. Corrosion Studies

a. Ferrous Alloys in Superheated Steam. A refreshed autoclave test of Types 304 and 406 steels in oxygenated (30-ppm) steam at 600°C and 42 kg/cm² was completed. The results are shown in Figure 18. The corrosion rate of Type 304 steel is estimated at about $2\frac{1}{2}$ mdd, about half of the value previously measured for 650°C. Also, the total corrosion at 70 days was less than half that measured at 650°C for a similar period. Type 406 steel rapidly corroded to about 3 mg/cm² with very little subsequent corrosion. This behavior is similar to that previously observed at 650°C.

Figure 18. Corrosion of Stainless Steels in Oxygenated Steam (600°C - 42g/cm²)

Currently a test in oxygenated (30-ppm) steam at 650°C and 7 kg/cm² is being performed to determine the effect of pressure on corrosion rate for Types 304 and 406. After exposure for 2 and 4 weeks, the behavior of Type 304 (electropolished) is quite similar to that noted for this alloy in a test run at 42 kg/cm². Type 406, however, corrodes less at the lower pressure. Portions of the Type 406 specimens are still covered with a thin temper film after four weeks of exposure.

2. Ceramic Fuels

a. The Uranium-Plutonium Carbide System

(i) Preparation. Two methods are being investigated for the production of solid-solution uranium-plutonium carbides in the range from (U_{0.9}-Pu_{0.1})C to (U_{0.5}-Pu_{0.5})C. These are (1) the carburization of uranium-plutonium alloy, and (2) the combination of individually produced uranium monocarbide and plutonium monocarbide. Both melting and diffusion are being tried.

Previous work has shown that U-20 w/o Pu alloy is rapidly attacked by air and by the oxygen residual in the glovebox atmosphere. A 250-g batch of this alloy, carburized at 1700°C for 25 min, produced a carbon content of 1.9 w/o. A similar batch carburized at 1800°C for 20 min yielded a carbon content of 2.4 w/o. The resulting materials were heated to 2000°C and did not melt. The resulting carbon content was 3.8 w/o. The metallographic samples were rapidly attacked making microscopic examination extremely difficult.

The first tests had been made on the diffusion combinations of uranium monocarbide and plutonium monocarbide. This was done by mixing 50-g batches of minus 80-plus 325 mesh powders and firing them at 1900°C for a period of 10 min in a previously carburized tantalum crucible. A fused cake resulted. Metallography indicated that most of the uranium carbide particles were encased in plutonium carbide. There was little evidence of diffusion. These cakes were then refired at 2000°C for 10 min. The microstructures show shaded diffusion zones with smaller amounts of PuC still remaining in the grain boundaries. Additional experiments are in progress to determine temperature, time, and particle-size relationships for producing a uniform solid solution.

Preparation of an additional 400-g batch of uranium carbide has been completed by the usual method of adding carbon to uranium dissolved in zinc-14 to 18 percent magnesium solution. The carbide had the lowest oxygen content (0.29 w/o) achieved in this series of runs. This is attributed to better degassing of the activated charcoal that is used as the carbon source and to improved handling procedures. The carbon content of the product was 5.56 w/o (the theoretical carbon content of uranium monocarbide is 4.8 w/o).

(ii) Irradiation Studies. Two specimens of vibratory-compacted PuC powder and four specimens of vibratory-compacted physical mixtures of UC-20 w/o PuC powders have been examined after irradiation in an instrumented capsule in the CP-5 reactor. Burnups ranged from 5.0×10^{20} to 7.1×10^{20} fiss/cc (17,500 to 26,200 MWd/MT carbide). Mean cladding surface temperatures for the irradiation period were from 370 to 515°C, the range of maximum cladding temperatures being from 405 to 625°C.

The specimens were jacketed with 0.23-mm (0.009-in.)-wall Type 304 stainless steel or Nb-1 w/o Zr with a 4.42-mm (0.174-in.) O.D. The fuel was compacted to approximately 80% of theoretical density in a 50-mm (2-in.) length with a 13-mm (0.5-in.)-long helium-filled reservoir left above the fuel. Size ratios of the powders in all specimens were 60 w/o of 20/40 mesh, 20 w/o of 120/200 mesh, and 20 w/o of -325 mesh. The PuC content in the mixed carbide specimens made up the entire 120/200 mesh fraction. Carbon contents in the six specimens ranged from 3.40 to 5.14 w/o C.

Neutron radiographs of the specimens while still encapsulated showed that (1) no cladding failures had occurred and the fuel was completely contained, (2) the fuel did not appear to change in length, (3) the mixed UC-PuC powders were inhomogeneous after irradiation either from failure to form a solid solution or from induced segregation, and (4) a thin, irregular central void may exist in at least two of the UC-PuC specimens.

Visual examination when the pins were removed from the irradiation capsule verified the neutron-radiograph information that none of the specimens had failed. The Nb-1 w/o Zr claddings were covered to varying degrees with black coatings believed to be an oxide of niobium. The stainless claddings were comparatively bright. Diameter measurements showed cladding changes no greater than 0.05 mm (0.002 in.) or 1%. Volume changes measured by immersion were of the order of 0.5% for UC-PuC pins and up to 1.1% for the PuC pins. The magnitude of these changes, however, is too small to make definite comparisons between PuC and UC-PuC specimens, or between stainless steel and Nb-1 w/o Zr claddings.

The specimens were punctured for determination of fission gas-release rates. Measured bulk gas release from the specimens roughly correlates with the burnups and irradiation temperatures, with the exception of two UC-20 w/o PuC pins, which released less gas by factors of five and ten to one. The PuC used in both of these pins had been remelted in fabrication prior to crushing; this could have resulted in less absorbed gas in these specimens. The gas samples are presently being analyzed mass spectrometrically for all constituents. Maximum calculated internal pressure in the gas reservoir of the hottest specimen, clad with Nb-1 w/o Zr, was 20.63 kg/cm² (293 psi), or a cladding hoop tensile stress of 207.1 kg/cm² (2800 psi).

b. Properties of (Th-U-Pu) Phosphide. As part of the program on (Th-U-Pu) phosphide bodies, studies of the oxidation of UP powder are continuing. The powder is being exposed to static air at low temperatures (200-270°C) to gain information on the effect of moderate oxidation on the UP structure. Temperatures not exceeding 300°C are being employed during the first stages of oxidation to avoid ignition of the powder.

The UP structure showed signs of degradation with an oxygen pickup of 2%. The high-angle X-reflections became diffuse, and the lattice showed a moderate increase in size. These two phenomena may be related to the occurrence of oxygen atoms in interstitial positions, producing an enlarged and strained lattice. This trend continued when the oxygen content was increased to 4%: the structure had deteriorated further and the lattice had become slightly larger. At this point another crystalline phase, the higher uranium phosphide, UP_2 (small amounts of U_3P_4 may also have been present), was observed. Apparently, during the early stages of oxidation, oxygen atoms displace or otherwise eject phosphorus atoms from the rock-salt structure, which then react with UP to form the higher phosphide. Therefore, it may be deduced that the oxygen atoms are attracted preferentially to uranium atoms and that UP_2 has greater oxidation resistance than UP.

The UP structure exhibited no further breakdown when the oxygen content was increased to 6%, at which stage UO_2 was first observed. Apparently, after some point no further oxygen can be accommodated interstitially in the UP structure and formation of UO_2 then begins. More UP_2 was observed in material containing an oxygen content of 6% as compared with that with a 4% oxygen content. Except for increased amounts of UP_2 and UO_2 , material with an oxygen content of 8% was similar to that containing 6% oxygen.

3. Particulate Metal Fuel Elements

a. Vibratory Compaction. A device to impart a horizontal mode of vibration to specimens simultaneously with vertical vibration has been developed. The bridging condition previously reported was alleviated by the use of this device. However, during compaction separations of the binary fractions took place with the larger fractions being displaced vertically. The erratic results produced by separation of the fractions was corrected by loading the second size fractions through a screen with apertures small enough to hold down the larger-sized components and to allow the second fraction to pass through into the body of the larger fraction. Results of experimentation with the second of the two methods outlined above are quite consistent and thereby lend themselves to more thorough analysis.

b. U-Pu-Fz Specimens. Specimens were made with powdered U-15 w/o Pu-10 w/o Fz alloy. The powders were produced by micropulverizing injection-cast pin shards. The powders were screened into minus 325 mesh, minus 200-plus 325 mesh, and minus 120-plus 200 mesh size fractions. Three specimens of each size fraction were prepared by vibratory compacting into 8.26-cm (3.25-in.)-long by 3.96-mm (0.156-in.)-inside diameter Nb-1 w/o Zr tubes, which were then closed. The powder packing densities varied between 55 and 60 percent. The TIG welding method was used to seal the specimens, which were leak detected and also were examined radiographically. The X rays showed a lower density near the top of the fuel column than at the bottom. Fixed contamination of the surfaces varied from less than 100 to 1100 dpm in the weld areas. The samples were then cleaned until no counts appeared on filter paper wipes.

4. Extrusion Developments

a. Production of Mo-0.5 w/o Ti-0.08 w/o Zr (TZM) Tube Blanks. Three TZM sleeves have been extruded by a technique that used a Type 304 stainless steel jacket and core. One DPM billet had an Al_2O_3 -filled annulus between the TZM sleeve and the Type 304 stainless steel core. Reduction ratios ranged from 7.2 to 9.9:1, and the extrusion temperature was dropped from 1150°C for the first billet to 1100°C for the last two billets at 9.9:1 reductions. Radiography showed the TZM sleeves to be uniform. Further fabrication of the TZM tube blanks will follow complete evaluation of the extruded product.

5. Physical Properties of Uranium and Plutonium

Preliminary studies of the critical properties of uranium and plutonium, using the methods suggested by Grosse,⁴ have yielded the following values for the critical temperatures, densities, pressures, and compressibilities:

- (a) For uranium $T_c = 12,500^\circ\text{K}$; $D_c = 3.22 \text{ g/cm}^3$; $P_c = 6,860 \text{ atm}$; $Z_c = (P_c M)/(RT_c D_c) = 0.490$.
- (b) For plutonium $T_c = 10,000^\circ\text{K}$; $D_c = 1.41 \text{ gm/cc}$; $P_c = 1,870 \text{ atm}$; $Z_c = 0.387$.

The value of critical temperature of uranium agrees well with that estimated by Grosse, but is about 14% or more below that estimated by Brout,⁵ who used a principle of corresponding states based on a critical

⁴A. V. Grosse, The Temperature Range of Liquid Metals and an Estimate of Their Critical Constants, J. Inorganic and Nuclear Chem., 22, 23 (1961).

⁵Robert H. Brout, Equation of State and Heat Content of Uranium, APDA-118 (Feb 1957).

compressibility factor $Z_c = 0.30$ and other reduced quantities. The constants for plutonium have not been determined previously.

The calculated values of Z_c are noticeably higher than the values 0.27-0.30 for ordinary fluids. For mercury, the only metal for which experimental critical properties are available, $Z_c = 0.475$. Therefore, simple application of corresponding states can be seriously in error for the metallic state.

However, the ratio of the critical temperature to the normal boiling temperature, T_c/T_b , which is about 1.5 for normal fluids, is 2.75 for mercury and is estimated as 2.86 for both uranium and plutonium. This may yield a simple rule of thumb for estimating critical properties of metals when data are unavailable.

Work is in progress to get an independent estimation of critical properties for uranium and plutonium by the method of Eyring.⁶ This will also lead to estimation of thermodynamic and transport properties and their pressure variation.

An additional method being studied is to estimate constants for an intermolecular potential and to use this to get virial coefficients and transport integrals for calculating the thermodynamic and transport properties. Tests of this against existing metal vapor data and quantum-mechanical calculations indicate collision cross sections are smaller than would be estimated by existing techniques.

6. Nondestructive Testing

a. Neutron Imaging. The use of uranium screens for both direct exposure and transfer techniques has been studied. Uranium metal screens of various U^{235} enrichments, each with a thickness of the order of 0.04 mm, were used. In both exposure techniques the screens with higher enrichment yielded the fastest results. A uranium screen having an enrichment of 93% produces a relatively fast single-screen direct-exposure result for either front or back films. The speed result was only slightly slower than the single gadolinium-screen direct-exposure method. However, the resolution result with the uranium screen did not approach those obtained from use of gadolinium screens.

As a transfer screen, the 93% enriched uranium was somewhat faster than with the use of indium and appeared to yield a comparable resolution. The radioactivity of the enriched uranium screen was a mixture of several activities having different half-lives. However, the activity of the screen appeared to decay to about 10% of the peak value within 2 hr. A 2-hr transfer time was employed.

⁶Charles M. Carlson, Henry Eyring, and Taikyue Ree, Structures in Liquids. V. Thermodynamic and Transport Properties of Molten Metals, Proc. Nat. Acad. Sci., USA, **46**, 649 (1960).

Application work completed during this period includes the inspection of one irradiated reactor fuel capsule and the radiographic inspection of 180 cm of 7.5-cm-diameter uranium-molybdenum alloy rods.

b. Ultrasonic Imaging. The resolution study involving the continuously pumped ultrasonic image system has been extended to lens systems with the aluminum test objects having holes all the way through. Results were, in some cases, difficult to interpret, probably because of diffraction effects introduced by the single plano-concave plastic lens itself. However, depth of focus was of the order of 2.5 cm on either side of the optimum setting. A magnification of about 2X was used for these tests. The hole sizes detectable did not differ appreciably from those previously reported for the direct shadow technique.

Some studies of the angular response of the system indicate that the camera responds well to ultrasonic radiation striking the piezoelectric target at angles as high as 15° from the normal. At larger angles, the image begins to degrade appreciably.

Other angular studies have indicated that flat, nonbond test objects should be angled as much as 10° to 20° from the normal for best imaging results. This appears to be true because there is then less tendency for standing waves, which interfere with the useful signal, to be set up between the detector and the test object.

Resolution studies with aluminum test plates containing flat-bottomed holes, drilled part-way through, are still in progress.

c. Correlation of Heat Transfer Properties and Bond Quality. Upon the completion of the flash tube apparatus for determining thermal diffusivity and thermal conductivity, the system was checked by measuring the thermal diffusivities of several different metals, with the following results:

Specimen	$\alpha(\text{cm}^2/\text{sec})$		Percentage of Difference
	Measured	Literature*	
Brass	0.337	0.339	0.6
Aluminum (2S)	0.872	0.860	1.4
Nickel (A)	0.154	0.155	0.64
Steel (1018)	0.1647	-	-
Cadmium	0.450	0.467	3.6
Copper	1.11	1.14	2.6

*Values taken from Handbook of Physics

Several bonded specimens consisting of two 0.063-cm-thick pieces of copper were bonded with soft solder. Values of the diffusivity ranged from $1.11 \text{ cm}^2/\text{sec}$ for solid copper to $0.063 \text{ cm}^2/\text{sec}$ for a specimen that contained a complete air gap; the most uniformly well bonded specimen that could be made gave values from 0.669 to $0.76 \text{ cm}^2/\text{sec}$. For any other specimen, the values of diffusivity were found to vary within the above range with the region of the specimen that the thermocouple contacted, indicating that the method is very sensitive to bond variations over small regions (1-2 mm).

The physical setup is, at present, being modified to insure greater stability, a necessity for the thermal conductivity measurements that are to be made on the bonded specimens.

d. Application of Infrared Radiation. Investigation of the possible use of infrared in nondestructive testing has continued. Some additional tests were conducted with thermographic phosphor screens in the demountable ultrasonic imaging system to determine the feasibility of developing an electronically scanned infrared imaging system with this material. The results of these investigations indicated that when used in this manner the phosphor is primarily sensitive to visible light.

D. Reactor Materials Development

1. Structural Materials

a. Brittle Fracture Studies. Notch-impact tests on steels are being performed on an Amsler-Schnadt Impact Pendulum machine. This machine can be used for making standard Charpy V-notch tests and also for making Schnadt coheracy impact tests. The most important feature of the machine is the availability of two impact velocities which differ by about a factor of 50. The high speed is standard for Charpy tests.

(1) Calibration. The Amsler-Schnadt impact machine was calibrated with 15 Watertown Arsenal standard specimens. These specimens represented three energy levels: $12.5 \pm 1 \text{ ft-lb}$ ($1.73 \pm 0.14 \text{ kg-m}$); $44.5 \pm 2 \text{ ft-lb}$ ($6.15 \pm 0.28 \text{ kg-m}$); and $76.0 \pm 4 \text{ ft-lb}$ ($10.5 \pm 5.5 \text{ kg-m}$) at -40°C . The results of these calibration tests are given in Table XII, which shows individual values as well as the average obtained for each energy level. The following conclusions were reached:

(a) Within each energy level, the scatter was more pronounced as the energy level increased.

(b) The use of spring centering devices did not appear to influence the results.

(c) At the low and intermediate energy levels, the Amsler-Schnadt impact machine gave acceptable values, although the average in both cases was at the high end of the permissible range.

Table XII. Impact-energy Measurements Made with the Watertown Arsenal Standard Specimens - First Set

Documented Energy Level					
12.5 ± 1 ft-lb (1.73 ± 0.14 kg-m)		44.5 ± 2 ft-lb (6.15 ± 0.28 kg-m)		76.0 ± 4 ft-lb (10.5 ± 0.55 kg-m)	
kg-m	ft-lb	kg-m	ft-lb	kg-m	ft-lb
1.85	13.4	7.15	51.7	11.85	85.7
1.85	13.4	6.30	45.6	11.85	85.7
1.80	13.0*	6.25	45.2	12.25	88.6
1.85	13.4*	6.20	44.8	12.60	91.1
1.90	13.7*	6.40	46.3*	12.10	87.5*
Avg:	1.85	6.46	46.7	12.13	87.7

*Indicates specimens tested using spring centering device.

At the high energy level, however, the machine indicated an average which was appreciably higher than the standard value. Examination of the broken specimens revealed appreciable brinelling at the supports, which increased in intensity as the energy level increased. It was concluded that this brinelling action was the factor contributing to the high-energy readings and was due to the shape of anvil supports used on the Amsler-Schnadt machine.

In an attempt to correct the situation, a new set of anvils, meeting Watertown Arsenal specifications, was obtained. These are shown schematically in Figure 19.

A. SCHNADT ANVIL SUPPORTS

Figure 19

Schematic of Anvil Supports

B. WATERTOWN ARSENAL ANVIL SUPPORTS

A second set of standard specimens was procured from Watertown Arsenal. The results of these tests are shown in Table XIII. The measured impact energies fall within the prescribed limits. The machine is now considered to be a calibrated facility suitable for tests under the AEC Program on Radiation Effects in Structural Materials.

Table XIII. Impact-energy Measurements Made with the Watertown Arsenal Standard Specimens - Second Set

Documented Energy Level					
12.5 ± 1 ft-lb (1.73 ± 0.14 kg-m)		44.5 ± 2 ft-lb (6.15 ± 0.28 kg-m)		76.0 ± 4 ft-lb (10.5 ± 0.55 kg-m)	
kg-m	ft-lb	kg-m	ft-lb	kg-m	ft-lb
1.50	10.8	6.57	47.5	11.1	80.5
1.70	12.3	7.10	51.2	10.5	76.0
1.85	13.4	6.20	44.7	11.08	80.1
1.70*	12.3	5.90	42.7	11.0	79.5
1.65*	11.9	6.08	44.0	11.0	79.5
Avg:	1.69	6.37	46.0	10.95	79.1

*Indicates specimens tested using spring centering device.

(2) Preliminary Test Results. A set of 24 coheracy specimens was prepared from A-212B transverse blanks. Despite some dimensional inconsistencies, the specimens were adequate for preliminary investigations. The specimens were tested at two different values of impact velocity. A series of 19 standard Charpy V-notch transverse specimens made of A212B material was tested at the two impact speeds. The results indicate two distinct transition curves are obtained and are shown in Fig. 20

Figure 20. Comparison of Ductile-Brittle Transition Curves

in comparison with the curves obtained using the Schnadt coheracy specimens. As in the case of the coheracy tests, the results of the standard Charpy V-notch tests indicate a pronounced effect due to the impact velocity and temperature. Thus, for a given temperature and speed of testing, the value of specific impact energy was higher for the Charpy than for the Schnadt coheracy specimens. This behavior may be attributed to size effects as well as the existence of a compression zone in the Charpy specimen which is eliminated in the Schnadt coheracy specimen.

However, the significant feature of the comparison shown in Figure 20 is that the Charpy transition temperature is considerably lower than the Schnadt transition temperature for each of the two given speeds of testing.

2. High-temperature Mechanical Properties

Installation of an Instron Universal Tensile Testing Machine with an accompanying Brew high-temperature, high-vacuum furnace assembly has been completed. This equipment will be used for the measurement of mechanical properties of materials at elevated temperatures. A Riehle Model DV vacuum extensometer has recently been added to this equipment for making static elastic-modulus and yield-point determinations.

Initial mechanical-property measurements were made with Nb-1 w/o Zr alloy in the stress-relieved condition. The threaded-rod tensile specimen used had a 6.35-mm (0.25-in.) reduced diameter corresponding to ASTM standards. Tests were conducted in 5×10^{-6} mm Hg vacuums with a constant crosshead speed of 8.3×10^{-4} cm/sec (0.020 in./min). Results obtained to date are given in Table XIV.

Table XIV. Some Tensile Properties of Nb-1 w/o Zr

Temp (°C)	Ultimate Tensile Strength (10^3 kg/cm ²)	Yield Strength ^a (10^3 kg/cm ²)	Total Elongation (%)	Reduction of Area (%)	Elastic Modulus ^b (10^6 kg/cm ²)
300	5.32	4.74	28.1 ^c	90	1.24
642	4.34	4.20	37.5 ^c	93	0.84
850	3.62	3.44 ^d	14.4 ^e	87	f
955	3.59	3.48 ^d	15.2 ^e	90	f

^a0.002 strain offset.

^bStatic tangent elastic modulus.

^cMeasured in 2.54-cm (1-in.) gauge length.

^dMeasured from crosshead travel and may not correspond to true 0.002 offset yield strength.

^eMeasured in 3.16-cm (1.25-in.) reduced section length.

^fNot measured in absence of extensometer.

E. The Metallurgy Alpha-Gamma Hot Cell

Operation of the alpha-gamma hot cell in Building 212 continued on a limited basis pending completion of leak-rate tests and completion of the air-purge ventilation system for the service areas of the cell.

Six irradiated U-Pu-Fs alloy pins (previously removed from capsule No. CP-30 in Building 301 caves) were transferred into the alpha-gamma cell for density and dimensional measurements, and for visual examination. This material was the first irradiated plutonium introduced into the cell. Alpha contamination from the operation was negligible due to the integrity of the jacketed pins.

On two occasions technicians entered the gas cell, for less than 15 min, to correct or modify equipment installations. Scott Air Pak breathing air systems were used. The large lock was not purged to effect these entries; however, it was found that oxygen pickup could be held to less than 1% by closing the lock doors behind the men in the cell.

The gas cell was tested for in-leakage of air by valving off the nitrogen cell purge supply and noting the rise in the oxygen content over a 24-hr period. The cell was maintained at the operating pressure of -13 mm wc during this test. Air leakage was found to be 0.3 cc/sec. From the previously determined total in-leakage, it is concluded that 88% of the in-leakage is nitrogen from the pressurized seal annuli.

F. Heat Engineering Research

1. Heat Transfer in Double-pipe Heat Exchangers

Heat transfer between liquid metals in countercurrent flow, double-pipe heat exchangers has been studied further. The available mathematical techniques suitable for the cocurrent flow cannot be applied to the countercurrent flow as now formulated. Either more tractable formulations and/or new mathematical techniques are being sought.

As a means of comparing cocurrent and countercurrent heat exchanger operation without the necessity of finding a successful solution to the mathematical problem mentioned above, the analysis was simplified to make it possible to obtain analytical solutions for both types of operation. This simplification is the assumption of a uniform heat transfer coefficient for one side of the exchanger, and results in a mathematical formulation which can be thought of as representing heat exchanger between a liquid metal and a fluid of moderate Prandtl number.

The dependence of fully developed liquid metal heat transfer coefficients on exchanger operation has been studied by the simplified procedure. With cocurrent flow, the results are similar to those obtained previously for heat exchange between liquid metals (e.g., see Monthly Report for October 1962, ANL-6635): values of fully developed heat transfer coefficients can be significantly less than those corresponding to the boundary condition of uniform wall temperature, but never larger than those corresponding to the boundary condition of uniform wall flux. The results for countercurrent flow, however, are quite different: values of fully developed heat transfer coefficients can be significantly larger than those corresponding to boundary condition of uniform wall flux, but never less than those corresponding to the boundary condition of uniform wall temperature.

The results for both types of operation are illustrated in Figure 21, which applies to a double-pipe heat exchanger consisting of adjacent, infinitely wide, parallel plane flow channels separated by a common wall. In this figure, fully developed heat transfer coefficients, normalized with

Figure 21. Heat Transfer in Double-pipe Heat Exchanger

respect to values appropriate to the boundary condition of uniform wall heat flux, are plotted vs the heat capacity flow rate ratio H ; the parameter K_T is the sum of the thermal resistance ratios K and K_W :

$$\begin{aligned} H &= C_2 W_2 / C_1 W_1 & ; & & K &= k_1 k_1^+ / a_1 h_2 & ; \\ K_W &= k_1 k_1^+ b / a_1 k_w & ; & & K_T &= K + K_W & , \end{aligned}$$

where

$$\begin{aligned} C_i &= \text{specific heat of fluid in channel "i," Btu/(lb}_m\text{)(}^\circ\text{F)} \\ W_i &= \text{mass flow rate of fluid in channel "i," lb}_m\text{/hr} \\ k_i &= \text{thermal conductivity of fluid in channel "i," Btu/(hr)(ft)(}^\circ\text{F)} \\ a_i &= \text{width of channel "i," ft} \\ h_2 &= \text{heat transfer coefficient for channel "2" (assumed uniform} \\ &\quad \text{along channel length), Btu/(hr)(ft}^2\text{)(}^\circ\text{F)} \\ b &= \text{thickness of common wall separating flow channels, ft} \\ k_w &= \text{thermal conductivity of common wall separating flow} \\ &\quad \text{channels, Btu/(hr)(ft)(}^\circ\text{F)} \end{aligned}$$

In the above $i = 1$ or 2 . Channel "1" identifies that channel through which liquid metal flows and for which fully developed heat transfer coefficients are shown on the graph. The quantity k_1^+ is a dimensionless factor that accounts approximately for turbulent flow in channel "1" and is mainly a function of Peclet number. The relationship

$$k_1^+ \approx 0.97 + 0.0033 \text{ Pe}_1^{0.8} \quad ,$$

with Pe_1 representing the Peclet number for channel "1," gives sufficiently accurate results when $\text{Pe}_1 < 1000$.

Similar results have also been obtained for the concentric tube double pipe heat exchanger.

2. Natural-convection Heat Transfer in Liquid Metals

A study of natural-convection heat transfer rates in liquid metals is in progress. The equipment has been completely tested, and preliminary studies with water or mercury as the heat transfer fluid have been performed. For the calculation of temperature and velocity profiles, theoretical relationships have been developed which take into account the effect of thermal conductivity in the direction of convective flow. Theoretical curves have been developed for Prandtl numbers which cover a range of properties from air to liquid metal. Experimental data obtained with

water as the heat-transfer medium agree with the predicted values. The temperature profiles obtained with mercury flowing in natural convection up a heated plate are slightly flatter than predicted by theory.

G. Chemical Separations

1. Chemistry of Liquid Metals

a. Technetium-Zinc System. Studies of the technetium-zinc system by means of the recording effusion balance were continued. The decomposition pressure of $\text{Tc}_2\text{Zn}_{11}$ between 470 and 570°C may be represented by the equation

$$\log p_{\text{mm}} = 9.853 - 8242 T^{-1} .$$

By extrapolation of this equation, the peritectic temperature for the inter-metallic compound was found to be 915°C, in fair agreement with the peritectic temperature of about 950°C obtained by thermal analysis (see Progress Report for August 1962, ANL-6610, p. 58).

b. Plutonium-Cadmium Galvanic Cell Studies. The thermodynamics of the plutonium-cadmium system has been studied by means of two galvanic cells having the configurations

and

(see Progress Reports for May and July 1963, ANL-6739, p. 49, and ANL-6764, p. 49). The composite data from these cells may be fitted to two equations:

$$(350-402^\circ\text{C}) E (\text{V}) = 0.6910 - 0.5607 \times 10^{-3}T \quad ;$$

$$(402-631^\circ\text{C}) E (\text{V}) = 0.4032 - 0.06083 \times 10^{-3}T - 0.2947 \times 10^{-6}T^2 .$$

The data indicate that the peritectic decomposition of PuCd_{11} into PuCd_6 and a liquid cadmium solution occurs at 402°C. This is in excellent agreement with the results of solubility studies of plutonium in cadmium, which place the peritectic transformation at 404°C.

c. Rare Earth-Cadmium Galvanic Cell Studies. Study of the thermodynamics of rare earth-cadmium systems by means of a galvanic cell has been resumed. In earlier experiments, the cerium-cadmium system was studied (see Progress Report for September 1960, ANL-6234, p. 63).

In the current study, the praseodymium-cadmium system is being investigated. It is hoped that the systematic study of the rare earth-cadmium systems will lead to a better understanding of the influence of atomic size on the thermodynamic properties of these systems and will allow correlations to be made of such data as the distribution coefficients in metal-salt systems.

The praseodymium-cadmium cell has been assembled. The electrodes consist of praseodymium and a praseodymium-cadmium alloy. The electrolyte is a dilute solution of PrCl_3 in LiCl-KCl eutectic. In preliminary measurements, lower emf's were obtained with the praseodymium-cadmium cell than with the cerium-cadmium cell.

d. Liquid Metal Distillation. Studies of the vaporization of mercury from a liquid pool without the formation of bubbles (nonturbulent vaporization) were continued. In the most recent work, the top 4 in. of a pool of mercury, 1 in. in diameter and 12 in. deep, was resistance heated, and a vertical traverse of the pool centerline was made with a special thermocouple under the condition of nonturbulent vaporization. A temperature gradient of $26^\circ\text{C}/\text{in.}$ was measured in a 0.12-in.-thick surface layer. This may be compared with earlier observations of gradients in the range of 60 to $100^\circ\text{C}/\text{in.}$ in a 0.17-in.-thick surface layer (see Progress Report for September 1963, ANL-6784, p. 50) of a $2\frac{5}{8}$ -in.-diameter, 3-in.-deep resistance-heated pool. The vaporization rate in the 1-in.-diameter pool was $890 \text{ lb}/(\text{hr})(\text{sq ft})$ as compared with $590 \text{ lb}/(\text{hr})(\text{sq ft})$ in the $2\frac{5}{8}$ -in.-diameter pool.

For the 1-in.-diameter pool, only 3 percent of the total heat flux that passed through the vaporizing surface could be accounted for by applying a molecular conduction process to the gradient measured and assuming that the centerline gradient existed over the entire surface. This is much less than the 10 percent value found for the $2\frac{5}{8}$ -in.-diameter pool and suggests that a greater proportion of the energy was transferred by convective flow in the pool of smaller diameter.

The pressure in the vapor space above the smaller-diameter mercury pool was measured in preliminary studies and was compared with the saturation pressure at the temperature of the vaporizing liquid surface. It was found that the vaporizing mercury surface was superheated 24 to 30°C above the saturation temperature and that there was an inverse relationship between the superheat value and the vaporization rate.

e. Plutonium Recovery Process. Development work has continued on the use of a ternary cadmium-zinc-magnesium alloy as a process medium for the recovery of plutonium-bearing fast reactor fuels. Such a medium may permit the use of lower temperatures and stainless steel

equipment. The solubilities of uranium in two alloys (Cd-13 a/o Zn-13 a/o Mg and Cd-20 a/o Zn-10 a/o Mg) have been measured. The solubility of uranium in Cd-13 a/o Zn-13 a/o Mg alloy in the temperature range from 380 to 650°C reaches a maximum of about 3 w/o at 600°C. The maximum uranium solubility found in the Cd-20 a/o Zn-10 a/o Mg alloy in the temperature range from 380 to 650°C is 4.4 w/o at approximately 650°C.

2. Fluidization and Volatility Separation Processes

a. Alpha Decomposition of Plutonium Hexafluoride. The effect of alpha-induced decomposition of plutonium hexafluoride is being investigated as a part of a fundamental study of the radiation behavior of plutonium hexafluoride. Additional data have been obtained on the rate of alpha decomposition of plutonium hexafluoride (see Progress Report for August 1963, ANL-6780, p. 43). The decomposition rates for plutonium hexafluoride were determined in runs which investigated the effect of the surface-to-volume ratio of the test spheres, of prefluorinated versus non-prefluorinated test spheres, of plutonium tetrafluoride which was present in the test spheres from previous alpha-decomposition rate runs, and of plutonium hexafluoride alone and in the presence of fluorine, oxygen, and nitrogen. The runs were carried out in nickel spheres (about 125 cc) which were filled with 100 mm Hg of plutonium hexafluoride and held at about 25°C for 75 days. The results of the runs are summarized in Table XV. Unless otherwise indicated in the table, the spheres were prefluorinated. In nickel spheres which were packed with nickel wool, the geometric surface to volume ratio was increased tenfold. The results show the following effects on the average decomposition rate of plutonium hexafluoride:

(1) A small, but real, decrease in the decomposition rates was indicated when the alpha-induced decomposition occurred in the presence of plutonium tetrafluoride.

(2) A significant increase in the rate was noted in spheres that were packed with nickel wool as compared with spheres that did not contain nickel wool, indicating some surface effect.

(3) An expected increase in rate was indicated in a non-prefluorinated nickel sphere.

(4) An expected decrease in the decomposition rate was found upon the addition of fluorine (see Progress Report for April 1963, ANL-6717, p. 53).

(5) A decrease in rate was shown upon the addition of oxygen.

On addition of nitrogen, 35, 36, 42, 47, and 54 percent of the plutonium hexafluoride decomposed in mixtures which contained 88, 119, 382, 1080, and 2670 mm Hg of nitrogen, respectively, as the additive. In previous runs with nitrogen as the additive (see ANL-6717, p. 53), nitrogen trifluoride (NF_3) was found.

Table XV. Alpha-induced Decomposition of Plutonium Hexafluoride

Sample Storage Vessels:		Nickel spheres of 125 ± 3 cc volume. Unless otherwise indicated, prefluorinated spheres were used.	
Storage Temperature:		$25 \pm 2^\circ\text{C}$.	
Decomposition Time:		About 75 days.	
Pressure of Plutonium Hexafluoride:		100 mm Hg.	
Second Gas Component		Average Rate of Decomposition (%/day)	Remarks
Gas	Pressure (mm Hg)		
None	0	0.191 ± 0.031	
None	0	0.140 ± 0.030	Spheres used in previous tests; initially contained PuF_4 from earlier decomposition of PuF_6 .
None	0	0.300 ± 0.030	Spheres packed with nickel wool; surface to volume ratio increased 10-fold.
None	0	0.220	Sphere not prefluorinated.
Fluorine	138	0.082	
Fluorine	395	0.127	
Fluorine	3610	0.054	
Oxygen	148	0.099	
Oxygen	430	0.095	
Oxygen	2610	0.113	

b. Stress-corrosion Testing of Alloys of Aluminum and of Titanium. The testing of samples of 7075 aluminum alloy and of B-120 VCA titanium alloy for stress corrosion in an atmosphere of plutonium hexafluoride (see Progress Report for September 1963, ANL-6784, p. 52) were continued. These tests were being conducted at the request of the Oak Ridge Diffusion Plant. The stressed samples (two of titanium and four of aluminum) were placed in a quartz vessel containing plutonium hexafluoride (52 mm Hg) and held at a temperature of $65 \pm 3^\circ\text{C}$ following a 15-hr period during which the temperature was 28°C . It was previously reported

that the two titanium alloy samples which were stress-loaded to $140,000 \pm 1,100$ psi had failed during the first 6 days of testing and that the aluminum alloy samples which were stress-loaded to $61,000 \pm 600$ psi were still intact after 21 days of testing. After an additional 9 days (a total of 30 days), the aluminum alloy samples were still intact. This completes the stress corrosion testing of the samples.

c. Oxidation-Fluorination Reaction of Clad Uranium Dioxide Pellets in a Single Reactor. The development of a fluid-bed oxidation-fluorination reaction scheme for the processing of stainless steel-clad uranium dioxide pellets is continuing in engineering-scale runs. In this scheme, there are two distinct reaction zones: the first is a bed of uranium dioxide pellets with alumina particles in the voids (oxidation zone); the second is a bed of alumina particles placed over the uranium dioxide pellet bed (fluorination zone). In the oxidation zone, U_3O_8 fines are produced by contacting the uranium dioxide pellets with an oxygen-nitrogen stream which also serves to fluidize the alumina particles. The U_3O_8 fines are transported from the lower zone by the oxygen-nitrogen gas stream and are reacted in the upper zone with fluorine, which is admitted to the fluidized bed of alumina just above the uranium dioxide pellet bed. The fluidized alumina serves as a heat-transfer medium.

A run was made which continued the evaluation of the effectiveness of the two-zone oxidation-fluorination scheme for the removal of uranium dioxide from stainless steel cladding (see Progress Report for July 1963, ANL-6764, p. 53). To simulate sheared pieces of clad fuel elements, 11.6 in. long by about $\frac{1}{2}$ -in. diameter, Type 304 stainless steel tubing with the ends left open was used to contain uranium dioxide pellets (27 pellets each). The wall thickness of the cladding was 0.020 in. The cladding of the fuel segments was perforated with a single line of holes, each about $\frac{1}{16}$ in. wide by $\frac{3}{16}$ in. long and spaced about $\frac{11}{32}$ in. between centers.

In this run, a fuel charge of twelve 11.6-in.-long fuel segments (3.25 kg uranium dioxide) and a 17-in.-deep fluid bed of fused alumina (6.1 kg) were used. The fused alumina (120 mesh, Norton Blue Label Alundum) was screened to remove the fines (-200 mesh) prior to use. The run was made under the following operating conditions. In the oxidation zone, the composition of the fluidizing gas was 87 percent oxygen in nitrogen at a total gas flow rate of 0.3 cu ft/min. In the fluorination zone, the concentration of the fluorine in the gas mixture was from 9.3 to 24.1 percent (dilution of the fluorine was obtained by recycling the process off-gas at a rate of 0.65 cu ft/min). The temperature in the oxidation and fluorination zones was 475°C . Gas pulsing of the fluidizing gases was used; the pulse duration was 0.04 sec, with a pulse frequency of 2 pulses/min at a gas manifold pressure of 20 psig. The uranium dioxide was completely removed from the perforated stainless steel cladding in a processing time

of 7.5 hr (once-through gas flow). The overall fluorine utilization efficiency was 72 percent. The stainless steel cladding split completely during the run, and no evidence of uranium dioxide residue inside the cladding was found. Ninety-six percent of the theoretical amount of uranium hexafluoride was collected during the first 6 hr of operation, corresponding to an average uranium hexafluoride production rate of 30 lb $\text{UF}_6/(\text{hr})(\text{sq ft})$ at a fluorine utilization efficiency of 88 percent. The values obtained during the first 6 hr of operation represent the maximum rate of production of uranium hexafluoride and the highest fluorine utilization efficiency yet achieved in two-zone decladding experiments.

d. Recovery of Uranium from Highly Enriched Uranium-alloy Fuels by Chlorination and Fluorination Steps. Studies of the feasibility of using the fluid-bed chlorination-fluorination scheme for the recovery of enriched uranium from uranium-alloy fuels were continued. Presently, tests are being carried out with uranium-Zircaloy fuels (see Progress Report for August 1963, ANL-6780, p. 45). In the current scheme, the fuel is first reacted with hydrogen chloride in a fluid bed of inert solids. The zirconium is thereby converted to the tetrachloride, and the uranium is converted to the trichloride. Since the hydrochlorination reaction is performed at temperatures above the sublimation point of zirconium tetrachloride (331°C and 1 atm), a separation of the zirconium tetrachloride from the uranium trichloride is achieved. The uranium trichloride is then fluorinated to the volatile uranium hexafluoride and is thus removed from the inert solids of the fluidized bed

The objectives of the present study are (a) to determine if minor constituents in Zircaloy-2 and simulated fission products added to the fluid bed would affect the retention of uranium by alumina, (b) to determine the disposition and distribution of these minor constituents and fission products during each step of the reaction cycle, and (c) to determine if the alumina bed would cake as a result of interruptions of feed gas streams (which would allow the alumina bed in the reactor to become static) and as a result of elevated bed temperatures (to 600°C).

A run was carried out in a $1\frac{1}{2}$ -in.-diameter nickel reactor. The charge in the run consisted of two 141-g uranium-Zircaloy-2 alloy subassemblies (1.7 w/o normal uranium), each subassembly consisting of three plates. In addition to the fuel subassemblies, 4.8 g (1.5 w/o of the reactor bed) of eleven simulated fission products were also charged to the fluid-bed reactor. The eleven fission products were strontium, yttrium, niobium, molybdenum, ruthenium, rhodium, antimony, tellurium, cesium, barium, and cerium. The fluid-bed reactor and the packed-bed filter each contained 320 g of fused alumina (Alundum, Type 38; Norton Company). The fluid bed was composed of 50 w/o each of -40 +60 mesh and -80 +100 mesh alumina particles; and the filter bed was composed entirely of -14 +20 mesh alumina particles.

The following reaction sequence was employed in the run: hydrochlorination, hydrofluorination, and fluorination. The hydrochlorination step was conducted at a fluid-bed temperature of 375°C for 9.3 hr. After the first 3.4 hr of hydrochlorination, the operation was interrupted for 21.3 hr to simulate an electrical power failure; during the interruption, the feed gas stream was stopped, thus causing the alumina in the reactor to become static. Within the first few hours of the interruption, the reactor was rapidly cooled to room temperature and was maintained at this temperature for 2 hr. The hydrochlorination step was then resumed for the remaining 5.9 hr of the reaction step. Following the hydrochlorination step, with the bed again in a static condition, the reactor was rapidly heated to 600°C and was maintained at this temperature for 4.5 hr. The hydrofluorination step was carried out at a fluid-bed temperature of 350°C over a one-hour period. This was then followed by two fluorination periods of 2 hr each at fluid-bed temperatures of 250°C and 500°C. Planned interruptions (similar to those occurring during the hydrochlorination step) were carried out for the hydrofluorination and fluorination steps.

The retention of uranium by the fused alumina in the fluid-bed reactor and the packed-bed filter was unaffected by the presence of the simulated fission products. The final uranium concentration in the alumina bed was less than 0.01 w/o (1.3 percent of the initial uranium charge). The average uranium loss through the packed-bed filter during hydrochlorination was 0.1% of that initially charged. In spite of the long interruptions, during which the fused alumina in the fluid-bed reactor was in a static condition for 180 hr at temperatures between 350°C and 600°C and for 10 hr at 600°C, no caking of the alumina occurred and no difficulty in refluidizing the alumina was encountered.

The relative distributions of most of the minor constituents in Zircaloy-2 and the fission products were in conformity with the distributions estimated from the volatilities of their higher-valent chlorides and fluorides. Major amounts of aluminum (79%), boron (84%), and tin (97%) volatilized during the hydrochlorination step. Minor amounts of silicon (14%), iron (22%), and manganese (36%) volatilized during the hydrochlorination step; the remaining amounts of these elements were retained by the fused alumina grain. Molybdenum volatilized throughout the reaction cycle (42% during the hydrochlorination step and 51.5% during the fluorination step). Almost all (99.1%) of the titanium volatilized during the fluorination step. Of the elements studied, eight were completely retained by the fused alumina grain: silver (99.7%), barium (99.7%), cerium (100%), cadmium (100%), cobalt (100%), strontium (96.4%), and yttrium (100%).

e. Pilot Plant-scale Demonstrations of the Recovery of Uranium from Enriched Uranium, Aluminum, and Zircaloy Alloy Fuels. A shakedown run has been completed in the pilot plant installed to demonstrate the fluid-bed fluoride volatility process for the recovery of uranium from enriched

uranium-alloy fuels (see Progress Report for September 1963, ANL-6784, p. 53). In this run, a 16-plate subassembly (17.8 kg) composed only of Zircaloy reacted completely with hydrogen chloride in only 8.25 hr at fluid-bed temperatures which ranged from 400 to 450°C. Simultaneous operation of the fluid-bed pyrohydrolysis column for converting zirconium tetrachloride to the oxide was satisfactory. Following the hydrochlorination step, hydrogen fluoride was passed through the system in order to test the hydrofluorination procedure. In general, the results of the shakedown run were found to be satisfactory.

f. Conversion of Uranium Hexafluoride-Plutonium Hexafluoride Mixtures to Uranium-Plutonium Oxides: Preparation of High-density Particles. Plans are being made to study the conversion of mixtures of uranium hexafluoride and plutonium hexafluoride to the oxide by reaction with steam and hydrogen in a 2-in.-diameter fluid-bed reactor.

Current investigations are being made with uranium hexafluoride alone in preparation for the study with the UF_6 - PuF_6 mixture, and are being carried out in a 3-in. diameter fluid-bed reactor. Recent work has investigated the effect of high uranium hexafluoride feed rate and bed depth on uranium dioxide particle density.

The data from current and previously reported runs show that uranium dioxide particle density varies inversely with uranium hexafluoride feed rate and directly with the uranium dioxide particle bed height. For 9-in.-deep beds (static height), uranium hexafluoride flow rates of about 25 and 50 g/min produced uranium dioxide particles with densities of 9.3 and 8.8 g/cc, respectively. For 17-in.-deep beds (static height), uranium hexafluoride flow rates of 25 and 50 g/cc produced higher density uranium dioxide particles, 9.6 and 9.1 g/cc, respectively. An increase in the production of uranium dioxide fines also accompanied the increase in bed height. This effect may be advantageous, since the fines can serve as seed particles.

3. General Chemistry and Chemical Engineering

a. Head-end Treatments for Refractory Fuels. Head-end processes for converting uranium monocarbide fuels into a form that is soluble in molten chloride salts are being investigated. Since later steps in the reprocessing scheme use magnesium to reduce the uranium compound to uranium metal, it is desirable that the uranium compound be nonoxygenated so that solid magnesium oxide does not appear in the salt. The reprocessing scheme envisions countercurrent contacting of molten salt and molten metal in a packed bed; therefore, a particulate solid in the system would be detrimental. A scouting experiment was performed in which uranium monocarbide pellets were dissolved in NaCl-KCl-MgCl_2 at 500 to 600°C by treatment with hydrogen chloride. This treatment apparently converted the uranium carbide to a U(IV) species and free carbon.

b. Mixing in Liquid Metals and Salts. Work has been done to correlate mixing power input with the rate of reduction of thorium dioxide and uranium oxide to their respective metals in molten salt-metal systems. Six runs have been performed on a 400-g thoria scale and two runs on a 275-g urania scale. Typical processing conditions were used for each material except that mixing conditions were varied. It was determined that reduction rate is affected by the position of the agitator blades relative to the metal-flux interface, the type of agitator blade (i.e., propeller, turbine, or flat), and the mixing power input. It appears that mixing for 4 hr at power inputs of 0.1 to 0.3 hp/kg of urania or thoria will be required in order to effect complete reduction.

c. Solubility of Magnesium Fluoride in Magnesium Chloride-Calcium Chloride. The solubility of magnesium fluoride over the full range of magnesium chloride-calcium chloride compositions is being determined at various temperatures. Partial results indicate that the temperature coefficient of the solubility is small and that 5.0 m/o concentration of magnesium fluoride in the 47.5 m/o MgCl_2 -47.5 m/o CaCl_2 -5 m/o MgF_2 skull reclamation flux is near the solubility limit at 750°C.

d. Analysis of Mixtures of Plutonium and Uranium Hexafluorides. The use of gas chromatography for the quantitative analysis of mixtures of plutonium hexafluoride and uranium hexafluoride is being investigated. Initial chromatographic column work was done without plutonium hexafluoride and has been concerned with evaluating separations involving mixtures of uranium hexafluoride, sulfur tetrafluoride (SF_4), and sulfur hexafluoride (SF_6). This work has been carried out using columns which were packed with Kel-F 10 on Teflon 6, with helium used as the carrier gas. It has been shown that plutonium hexafluoride is selectively reduced to the tetrafluoride by SF_4 (see Progress Report for August 1963, ANL-6780, p. 47). If plutonium hexafluoride and uranium hexafluoride cannot be separated efficiently in the chromatographic column, then use of the reaction of plutonium hexafluoride with SF_4 could be made. The data have shown that SF_4 and its reaction product SF_6 can be separated from each other and that the sensitivity of detection of these compounds is at least ten times that of uranium hexafluoride. Thus, very small quantities of SF_6 (and thus PuF_6) can be detected. A high-efficiency chromatographic column (50 w/o Kel-F 10 on Teflon 6) should be capable of resolving these components (i.e., SF_4 and SF_6). Low temperatures and flow rates should also enhance SF_4 and SF_6 separations. The temperature of the column should be slightly above the boiling point of the lowest boiling component; operation of the column at lower temperatures would cause the chromatographic peak for UF_6 to become very broad and make difficult the quantitative estimation for UF_6 . Further work is planned on the development of this method.

4. Calorimetry

Standard-state calculations have been completed for the combustions of niobium, tantalum, and ruthenium in fluorine. Preliminary values (kcal/mole) for the formation of the pentafluorides from the elements according to the equation

at 25°C are the following:

	<u>NbF₅</u>	<u>TaF₅</u>	<u>RuF₅</u>
$\Delta E_f^\circ = \Delta E_c^\circ$	-432.1	-453.6	-211.9
ΔH_f°	-433.5	-455.1	-213.4
ΔG_f°	-406.2	-	-

The "uncertainty interval," equal to twice the combined standard deviations arising from known sources, was of the order of a few tenths of one kcal/mole in each case. No values for the Gibbs energy are given for TaF₅ or RuF₅ because of the unavailability of entropy values at 25°C for these substances.

An additional series of ten combustions of zirconium diboride in fluorine (see Progress Report for November 1962, ANL-6658, p. 56) has been completed. Impurity concentrations in the sample material are being determined to allow correction of the heat of combustion value.

H. Plutonium Recycle Program

1. Plutonium Recycle Fuel

The Plutonium Recycle experimental core will contain three different fuel zones:

- (a) a central zone of 36 elements fueled with 1.5 w/o plutonium oxide and 98.5 w/o depleted uranium oxide;
- (b) an intermediate shim zone of 60 elements fueled with UO₂ enriched to 6 w/o U²³⁵;
- (c) a blanket zone of 58 elements fueled with natural UO₂.

All plutonium-bearing fuel rods are being prepared by General Electric Hanford.

All shim and blanket elements will be fabricated by United Nuclear Corporation. United Nuclear is also preparing the feasibility report for processing of the enriched uranium fuel.

United Nuclear Corporation has reported good results in the preparation of a rare earth oxide dispersion in UO_2 by preparing a master blend of high-fired rare earth oxide with UO_2 , which is then added to the ceramic-grade UO_2 . For one powder blend containing an average of 1357 ppm of Sm_2O_3 , a standard deviation of 39.4 ppm was reported. The analytical error was 16.5 ppm. The variation from a true homogeneous dispersion at a 95% confidence level was $\pm 3.4\%$. Pellets fabricated from this material were analyzed, and the maximum variation from top to bottom in any pellet was reported to be 3.9%, within the tolerance specified.

A comparison will be made between dispersions of this high-fired rare earth oxide in UO_2 and dispersions prepared by the Dynapak high-energy-rate forming process developed at Hanford. Two master blends, one containing 5 w/o Sm_2O_3 and the other containing 10 w/o Sm_2O_3 , were prepared by Hanford by the Dynapak process and delivered to United Nuclear Corporation to be ground, screened, and blended with ceramic-grade UO_2 . This method may be used if a more uniform dispersion is obtained.

2. Plutonium Recycle Control Rods

Replacement control rods for EBWR have been redesigned to eliminate the riveted transition section between the absorber section and the Zircaloy-2 follower section. This new design incorporates a tongue-and-groove joint which will eliminate the stress due to differential thermal expansion of these two materials.

Further examination of data obtained from failed Inconel rivets removed from EBWR control rods, as reported in the September Reactor Development Progress Report, ANL-6784, p. 57 indicates that the failure cannot be definitely ascribed to stress-corrosion cracking. There was extensive evidence of intergranular cracks and perhaps some transgranular cracks.

A total of eight new control rods will be fabricated containing 2 w/o boron enriched to 92% B^{10} in stainless steel in place of natural boron in stainless steel for the absorber section. One central hafnium oscillator rod will also be replaced.

The boron stainless steel for the control rods will be procured from commercial suppliers, who have been invited to bid on the fabrication. The boron stainless and hafnium will be furnished by the Laboratory to commercial suppliers for the fabrication of the control rods.

3. Hydrodynamics

The hydrodynamics calculations (see Progress Report for September 1963, ANL-6784, p. 55) correlating regional power and void have been carried through two PDQ-CHOPPED iterations. The regional average

voids for the zero-burnup plutonium zone, first shim zone, second shim zone, and uranium zone were found to be in the ratio of about 1.0, 1.0, 0.9, and 0.3, respectively. For the case of europium and samarium burnable poison contents of about 39 gm and 7 gm per shim element, respectively, the power is limited to about 40 MW based upon a one-dimensional solution, if the above ratio of voids is used.

At a burnup of 0.006 in the plutonium zone, the relative regional ratio of voids become 1.0, 1.4, 1.3, and 0.55, respectively. These new voids at the higher burnup would limit the power to less than 30 MW.

Doppler reactivity effects have been calculated for a cold core at zero and at 0.006 burnup, and for the cold, plutonium region only at zero burnup. For the complete core, at an energy input of 1000 MW-sec (for which a melting temperature of the oxide fuel is reached in the core), the resulting δk_{eff} values were about -1.0% and -0.75%, respectively, for zero and 0.006 burnup. For the plutonium region only, at zero burnup, an energy input of 320 MW-sec (again reaching a point of fuel melting) resulted in a δk_{eff} of about -0.8%.

The values of δk_{eff} versus energy input will be used to supply reactivity feedback information to RE-129 kinetics calculations.

The new eight-group cross-section sets have been derived by means of GAM-1. These sets have a group structure intended to display detail in the range of the delayed-neutron spectra and are designed to study both hot and cold systems.

The 1188/RE code has been recompiled for use in cylindrical geometry. This code was used with the hot eight-group set to calculate a β_{eff} of 0.00555 and a prompt neutron lifetime of 37.6 μsec for the hot operating system at zero burnup. These parameters have been used to calculate the inhour curve by the RP-130 code.

The DTK transport theory code has been used in slab geometry to evaluate the epithermal self-shielding in the control rods. The parameters of a four-group set, which will be used in the PDQ rod calculations, were adjusted to give agreement with a 21-group calculation for correctly predicting the ratio of epithermal rod absorptions to total system absorptions. For the 90% enriched boron rods, the epithermal absorption cross section must be reduced by about 35%, whereas for the case of natural boron control rods, a reduction of about 10% is required to obtain agreement with the detailed 21-group results.

The 20-GRAND program is being checked with the CDC-3600 computer by comparison with some earlier PDQ results. If the program proves

to be acceptable, further two-dimensional calculations will be done with this new code. The 20-GRAND program has a potential advantage over the PDQ code for providing a complete description of the scattering matrix as opposed to the PDQ limit of only allowing down scattering by one group. This limitation results in a reactivity underestimate in the case of the plutonium recycle core for the EBWR, because neutrons which should actually skip over the Pu^{240} resonance group are being incorrectly absorbed in that group.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Magnetohydrodynamics (MHD)

1. MHD Power Generation - Jet Pump Cycle

Flow studies of a jet-pump MHD system using water and steam to simulate a liquid-metal system continue. After installing a 100-psi steam system, initial operation of the model linear generator was attempted. Tests have shown that pumping of the liquid is actually obtained and that a steady flow is in fact possible. Preliminary estimates show that a velocity of 50 to 70 fps is obtained in the constricted part of the test section. This is lower than desired. More complete data will be obtained and evaluated to determine the reason for this condition. In addition, variation of nozzle position will be tried to see if optimum settings are possible.

2. MHD Power Generation - Flashing Cycle

Work is continuing on the ac generator with pulsed flow (see Progress Report for September 1963, ANL-6784, p. 61). A generator has been fabricated which has two ducts through the magnetic field, which will allow a continuous flow of liquid metal. A gas pulsator has been fabricated which will operate at higher frequencies than the previous model.

An effort has been made to mock up the generator with a rotating wheel. Various widths of conductors can be placed on the wheel and rotated through a magnetic field. One electrode is taken from a liquid brush on the axis of the wheel, and the other is a liquid brush on the edge of the wheel. This apparatus can be used to examine pulse shapes and voltages for various conductor widths and electrode connections.

3. MHD Duct Flow

In a continuing study of convective magnetohydrodynamic channel flow, the problem of unsteady MHD generator operation has been considered. It was found that under certain general conditions when the temperature field is of a certain general form, exact solutions of the governing conservation equations could be obtained. Although numerical results are not yet available, it can be shown that the operation of a constant-area generator depends upon at least eight parameters (for laminar, single-phase flow): thermal and magnetic Prandtl numbers, Hartmann number, Rayleigh number, energy-generation index, a dimensionless driving force, a dimensionless wall-temperature function, and a wall-conductance ratio.

B. Rocket Fuel Test Reactor (RFTR)

The results of a preliminary physics and heat-transfer calculation for the Rocket Fuel Test Reactor (RFTR) show that a test reactor having the following characteristics can be designed:

- (1) The spatial distribution of the flux, power, and the energy spectrum of the flux which exists in the fuel of the ANL rocket design can be simulated in the test section of the test reactor.
- (2) A value of 4 to 5 can be achieved for the ratio of the maximum power density in the test section to that in the test reactor. A test reactor which is limited to a maximum power density of $2 \text{ MW}/\ell$ can be used to evaluate the performance of a rocket fuel at $10 \text{ MW}/\ell$.
- (3) The test reactor produces a large amount of power. Improving the simulation in the test section and increasing the maximum power density ratio result in larger power production of the test reactors investigated in the preliminary study.
- (4) The radius of the test section can vary between 3 and 13 cm without causing large changes in the outer radius of the test reactor.
- (5) Because of the limited nature of the study, only one type of fuel element, that proposed in the ANL rocket design, was investigated in any detail. However, this does not preclude the possibility of testing other types of fuel elements which are designed to operate in a fast-neutron spectrum.

The size and, in turn, the power production of the test reactor can be reduced by softening the spectrum; however, power peaks result in the test section from the addition of the neutrons of lower energy. Analysis of the physics calculations suggests two means of reducing the power without producing the power peaks. One is the addition of a neutron absorber between the test section and the test reactor to filter out the neutrons of lower energy. The other is to design a test reactor with a hard spectrum near the test section and a softer spectrum at the outer radius. Both approaches to the problem are under study.

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal Oxidation and Ignition Studies

a. Effect of Moisture on Oxidation of Plutonium. A series of experiments has been started to determine the influence of moisture on the oxidation of plutonium. Although other investigators⁷⁻¹⁰ have noted a marked effect of moisture on the rate of oxidation of plutonium below 100°C, no systematic study of the effect of water has been carried out at higher temperatures.

Preliminary studies of the effect of moisture on the isothermal oxidation of plutonium at 190°C have been conducted with oxygen which contained controlled amounts of moisture from 20 to 500 ppm. Plutonium in the form of 1-cm cubes was used. The experiments have demonstrated that as little as 180 ppm moisture causes the oxidation of the plutonium to accelerate to a maximum rate of about 20 $\mu\text{g}/(\text{sq cm})(\text{min})$, whereas 20 ppm produces a maximum rate of only 1.5 $\mu\text{g}/(\text{sq cm})(\text{min})$.

In previous isothermal studies of plutonium (see Progress Report for January 1963, ANL-6683, p. 50), it was found that, between 140 and 200°C, the parabolic rate was followed by a much faster linear rate that showed no temperature dependence. It is now believed that this very rapid rate resulted from the presence of moisture in the oxygen used as the reactant in the earlier runs.

2. Metal-Water Reactions

Attempts to compute curves of temperature versus time and curves of the extent of reaction versus time for the reaction of uranium spheres with water were continued. Computations carried out with the IBM-704 assumed that a heated uranium particle was contacted with water instantaneously, thereby simulating conditions encountered in both condenser-discharge experiments (see ANL-6543, p. 182) and in experiments in which metal particles are heated by means of a laser beam. Current computations indicate that the results of condenser-discharge experiments carried

⁷Kolodney, M., Protection of Plutonium Against Atmospheric Oxidation, LA-314 (June 1945).

⁸Waber, J. T., Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, United Nations, New York, 6, 204 (1958).

⁹Sachmann, J. F., Plutonium 1960, Cleaver-Hume Press, London, 1961, p. 222.

¹⁰Sachmann, J. F., Nature 197, 587 (1963).

out with uranium in water at room temperature or in heated water may be described by applying a parabolic rate law and an activation energy of 30 kcal/mole. The deviation between the computed values and the experimental values appear to be within the experimental error incurred in the measurement of particle size.

Preliminary studies of the kinetics of the reaction of molybdenum with steam have been completed. These experiments are a part of a study that has been initiated to examine the suitability of high-melting metals (m.p. above 2000°C), such as molybdenum, tungsten, tantalum, vanadium, and niobium, for use in the fabrication of equipment for metal-water studies. This work is not only important from a practical point of view but is also important because it will add to the knowledge of the kinetics of metal-water reactions.

The specimens of molybdenum tested had a purity of 99.95% and were in the form of right cylinders, 5 mm in diameter by 10 mm long. The cylinders were heated by induction while exposed to flowing steam. The rate of reaction was determined by measuring the rate of hydrogen evolution. The reaction followed a linear rate law over the temperature range from 1100 to 1550°C. The quantity of hydrogen generated and the loss of weight of the molybdenum specimens indicate that MoO_3 is the predominant species formed during the reaction. The rates of reaction of molybdenum with steam at the higher temperatures of the range studied were too rapid to allow the use of molybdenum as a structural material for high-temperature studies of metal-steam reactions.

B. Fast Reactor Safety Studies

Two safety features characteristic of typical fast power reactor designs are the short prompt-neutron lifetime and the quantity of fissionable material, which is usually sufficient to provide a number of critical masses. Because of these features, a class of "meltdown" accidents can be postulated in which some operating abnormality triggers some degree of fuel failure, the fuel is moved into a more reactive configuration, and a destructive burst of nuclear energy results. However, the factors influencing fuel failure and movement, and the degree of coherence in movement are complex. They are being studied experimentally in reactor meltdown experiments conducted in the TREAT facility, and theoretically in related analyses.

1. Fast Reactor Analog

Previous studies of three hypothetical accidents in an EBR-II-like system (see Progress Report for September 1963, ANL-6784, p. 66) have been made by means of existing digital computer codes which did not couple kinetics and transient heat transfer calculations. A fast reactor

analog has been set up to couple heat-transfer calculations with intact fuel-element feedback to kinetics computations. When necessary, in order to make possible a study of several decades of reactor power, analog calculations are made with a logarithmic neutron density kinetics circuit coupled to the temperature calculation. Open-loop (constant power, with no feedback) computations have been made for comparison with the earlier ARGUS temperature calculations. Computations for both pump failure and coolant-orifice block were compared. In the former, coolant momentum causes flow to decay slowly with an exponential decrease having a time constant of the order of 10 sec. The latter case is essentially a static heat-transfer calculation with stagnant coolant. Both cases showed a more nearly linear temperature rise to the onset of melting in the analog calculations, but this can be related to a difference in specification of material properties over the temperature range from 600 to 1010°C; one averaged value of each property is given in the analog, but the ARGUS input has a phase change, with appreciable heat of transformation, at 630°C. Rates of temperature rise appeared to be sufficiently close between the two types of calculation so that the analog could be relied upon to give good estimates of the time spread in failure for calculations of meltdown accidents.

The dropped-subassembly case reported last month (see ANL-6784, p. 68), which was calculated on the basis of adiabatic heating of the fuel, has been re-run on the analog. The adiabatic assumption was confirmed as reasonable, but heating of coolant was found to be appreciable by the time the coolest fuel elements attained the threshold of failure. One significant difference, however, was the effect of the appreciable heat of fusion of the fuel, an item not included in the earlier kinetics calculation. If the effects of both heat transfer to coolant and heat of fusion of the uranium-fission alloy are included, the time spread in failure over the reactor is increased by about an order of magnitude over the earlier estimate of about 4 ms.

2. Analysis of Pressure Surges in Fast Reactor Coolant Channels

Approximate scaling laws have been derived for pressure and time of expulsion of coolant from core channels on the assumptions that mechanical effects and finite limits on temperature penetration into coolant can be ignored. Time for expulsion after coolant saturation conditions are reached is approximately proportional to

$$(m_0 C / A q)^{1/3} ,$$

whereas the maximum pressure generated during the expulsion is approximately proportional to

$$(q^2 / A C^2 m_0)^{1/3} .$$

In these expressions m_0 is the coolant mass to be accelerated, A is effective coolant cross-sectional area into which coolant vaporizes, q is power entering coolant, and C is the derivative of coolant pressure with respect to enthalpy along the saturation curve. (In general, this quantity is highly nonlinear; however, for the cases studied thus far, the important temperature range has been sufficiently short that C could be approximated as a constant.)

Calculations on the package sodium loop have continued. The first calculation (see ANL-6784, Progress Report for September, 1963, p. 68) was for a single EBR-II element inside a coolant channel equivalent to the channel per element in an EBR-II subassembly. A more realistic mockup has been made of the actual coolant environment and coolant cross section for one heated element inside a cluster of dummy elements. Pressure at the time of expulsion from the loop test section for a constant value of q , equivalent to that from a 4.1% enriched fuel element with TREAT reactor power of 1300 MW, was estimated to be about 5 atm, and the time of expulsion to be about 20 ms.

C. TREAT

1. Small TREAT Loop

As a matter of safety, the first flowing sodium loops are being equipped with a sodium leak detector which will automatically shut down the entire system in the event of sodium leakage from the loop. The method of detection depends upon a globule of sodium falling upon a wire mesh and shorting an electrical circuit thus scrambling the reactor.

A rough draft of the operating manual has been prepared and is in review.

2. Large TREAT Loop

To support the conclusions drawn from the thermal-series analysis report by the Franklin Institute (see Progress Report for September 1963, ANL-6784, p. 69), calculated skin stresses in selected regions of the large TREAT loop were compared with results obtained from similar investigations. It was found that Type 347 stainless steel which was subjected to comparable thermal stress conditions for over 10,000 cycles incurred no damage.

3. Experiments in Progress

The series of transient heating experiments with prototype Hallam Nuclear Power Facility uranium carbide fuel that was started last month was completed. The samples were returned to Atomics International for examination.

Five metal-water reaction samples were irradiated during the month. Two samples were zirconium-uranium alloy plates in room-temperature water; two were zirconium-uranium alloy plates in water at 285°C; and, one was a zirconium-uranium-niobium alloy fuel pin that had been previously irradiated to a burnup of 1.6% of the uranium-235.

Two pre-irradiated EBR-II fuel pins and two pre-irradiated Fermi-I fuel pins were subjected to transient irradiations in transparent capsules. This was the first series of experiments in which high-speed motion pictures were obtained of the failure of pre-irradiated samples.

VI. PUBLICATIONS

Papers

REACTOR SAFETY EXPERIMENTS

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CERTIFICATION OF ALGORITHM 210 - HERMITE INTERPOLATION

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THE SCATTERING OF FAST NEUTRONS FROM W¹⁸⁴

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H. C. Thacher, Jr.

Math. Computation 17(84), 485-486 (October 1963)FISSION CROSS SECTION OF Pu²³⁸ (Abstract)

D. K. Butler

The AEC Nuclear Cross Sections Advisory Group Meeting at the University of Colorado, August 13-14, 1963. WASH-1044, p. 5

GOOD RESOLUTION STUDIES OF ELASTIC NEUTRON SCATTERING FROM Fe (Abstract)

S. A. Cox

The AEC Nuclear Cross Sections Advisory Group Meeting at the University of Colorado, August 13-14, 1963. WASH-1044, p. 2

FAST NEUTRON CAPTURE CROSS SECTIONS (Abstract)

S. A. Cox

The AEC Nuclear Cross Sections Advisory Group Meeting at the University of Colorado, August 13-14, 1963. WASH-1044, p. 13

TIME-OF-FLIGHT MEASUREMENTS - DIFFERENTIAL ELASTIC
SCATTERING (Abstract)

Daniel Reitmann and A. B. Smith

The AEC Nuclear Cross Sections Advisory Group Meeting at the
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