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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:

August 1963	ANL-6780
September 1963	ANL-6784
October 1963	ANL-6801
November 1963	ANL-6808
December 1963	ANL-6810
January 1964	ANL-6840

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I. BOILING WATER REACTORS

A. BORAX-V

1. Operations

Preparations for operation of the peripheral superheater core were nearly completed. The fuel for the central superheater core was removed from the reactor vessel and placed in the storage pits, the control rod drives were disconnected, and the control rods were transferred to the storage pit for inspection. The central superheater core structure was removed and the peripheral superheater core structure was installed in the reactor vessel. Control rods and drives were reinstalled, and preparations for loading the core with the peripheral superheater were nearing completion.

Removal of the central superheater core structure was accomplished by semi-remote procedures, although its radioactivity level was low due to the short period of operation. The procedures, including the removal of flood-and-drain piping connections, appear to be satisfactory when radioactivity levels are high. Installation of the peripheral superheater core structure included connecting the flood-and-drain piping and radiographing of new welds.

The five cruciform and four T-shaped control rods were inspected. Their stainless steel-clad Boral poison sections passed the 0.330-in.-thickness gauge and the 0.390-in.-angularity gauge. Nominal control rod thickness is 0.312 in. Visual inspection of the poison section showed no rub marks or bulges.

The laminated X-8001 aluminum followers of the T-shaped control rods would not pass the 0.438-in.-thickness gauge or the 0.438-in.-angularity gauge. The 0.438-in.-angularity gauge would pass all of the followers on the cross control rods, and the 0.365-in.-thickness gauge would pass about half the followers. Closer inspection revealed that some of the spot welds holding the $\frac{1}{16}$ -in.-thick follower cladding to the $\frac{3}{16}$ -in.-thick core plate had broken, causing minor bulges in the follower section. Rub marks were apparent on the followers.

A stainless steel Type 304, countersunk screw-and-nut fastener, of the same type used to join the poison section to the follower, was used to repair the broken spot welds. To provide additional venting of the laminations, seven $\frac{1}{8}$ -in.-dia holes have been drilled through the cladding on each side of all follower blades.

All control rod followers passed the 0.365-in.-thickness gauge and the 0.390-in.-angularity gauge upon reinspection. These control rods are also within straightness-and-twist tolerance as determined by the straightness-and-twist fixture.

The shroud channels for the control rods of the central superheater core structure were inspected with a thickness gauge. An 0.467-in. thick gauge passed all channels except for control rod positions No. 3 and No. 5. An 0.441-in.-thick gauge passed these two channels. The reduced thicknesses of the channels are at points approximately 30 in. from the top of the shroud, or approximately 2 in. below the top of the core. An 0.485-in.-thickness gauge has passed all channels prior to operation.

The shroud channels for the control rods of the superheater core successfully passed 0.485-in.-thickness gauge.

The control rod drives were inspected. The linear floating ring labyrinth seals which contained some rust and fine particles were cleaned. The Teflon wipers for the extension shaft, although in good condition, were replaced. The chrome-plated 17-4 PH stainless steel extension shafts were in good condition with the exception of a superficial longitudinal scratch in the plating on one shaft. This shaft was stoned and reinstalled.

2. Experiments

a. Power Mapping of Core CSH-1. Data from the fine power mapping carried out at room temperature in central superheater core CSH-1 have been analyzed. In these experiments boric acid had been added to the reactor water to permit almost complete withdrawal of the control rods. Analysis was complicated by difficulty in normalizing between successive irradiations that were required to map one octant of the core and also by suspected irregularities in the counting equipment. Normalization was ultimately carried out by use of data from Plumlee *et al.*,¹ for the flux profile across a 4-plate superheater fuel element (perpendicular to the plates). Figure 1 shows the local-to-average ratio of power produced in the individual fuel assemblies in the core with eight bunched flow rods per boiling fuel assembly and with a drained superheater. The values for the superheater are low because a superheater fuel assembly contains 430 g of U^{235} as compared with 668 g of U^{235} in a boiling fuel assembly. About 17% of the power is produced in the superheater, compared with the 19% predicted for the reference design core (with 49 fuel rods per boiling fuel assembly).

b. Radiation Damage to W-W/26% Re Thermocouples. An experiment to determine the radiation sensitivity of W-W/26% Re thermocouples was carried out in the EBR-I reactor and hot cell in order to explain the temperature changes of several hundred degrees observed for boiler fuel rods during operation of BORAX-V boiling core B-2 (see Progress Report for March 1963, ANL-6705, p. 8).

¹K. E. Plumlee *et al.*, Critical Experiment with BORAX-V Internal Superheater, ANL-6691 (Nov 1963)

Legend:

⊗ Flow Rods

Notes:

Room Temperature

Superheater Drained

Boric Acid Concentration:

10.8 g(H_3BO_3)/gal (H_2O)

Figure 1. Local-to-average Ratio of Power Produced in Individual Fuel Assemblies of Central Superheater Core CSH-1 of BORAX-V

A pair of chromel-alumel thermocouples and one BORAX-V fuel rod thermocouple (W-W/26% Re) were installed in the fixture shown in Figure 2 and inserted in a thimble in the EBR-I core. Gamma and neutron heating of the fixture provided maximum temperatures of 690°F while in a fast flux of about 10^{14} nv. The thermocouples were exposed to six cycles of reactor startup and shutdown, and received a total fast neutron exposure of approximately 2×10^{18} nvt. During four steady-state periods of reactor operation at 975 kW, the W-W/26% Re thermocouple attained an indicated temperature of $665 \pm 5^\circ\text{F}$, while the two chromel-alumel control thermocouples indicated a temperature of $685 \pm 5^\circ\text{F}$. There was no apparent shift in calibration among the three thermocouples during the four maximum power cycles of reactor operation.

Figure 2. Device Used in Thermocouple Radiation-damage Experiment in EBR-I

To verify the findings that radiation had no apparent effect on the performance of the thermocouples, the three thermocouples and fixture were placed in a muffle furnace in a hot cell and calibrated at temperatures up to 1997°F. Three temperature cycles were run from room temperature to 1515°, 1880°, and 1997°F, respectively, before the test was terminated by failure of the tantalum-sheathed W-W/26% Re thermocouple due to loss of the argon purge gas supply. The calibration between the two chromel-alumel control thermocouples and the W-W/26% Re thermocouple showed a maximum disagreement of 22°F over the range of 500-1800°F, and there was no noticeable "drift" in calibration from one thermal cycle to another.

The tests showed that irradiation, for the conditions described, did not affect the performance of the W-W/26% Re thermocouple.

II. LIQUID-METAL-COOLED REACTORS

A. General Fast Reactor Physics

1. ZPR-III

Experiments with Assembly 44, the RAPSODIE mockup, continued. Following the initial construction of a clean, cylindrical critical loading (see Progress Report for January 1964, ANL-6840, p. 13), the engineering reactor configuration is being built by successive modifications to introduce axial blankets and transition zones between core and blanket (end gaps), which approximate the actual RAPSODIE design. The reactivity effects associated with these various engineering features of the design are being determined in the course of this step-wise rebuilding.

The reference critical composition given in Table VIII of the January Progress Report consisted of 32 core drawers in each half, loaded in a cylindrical configuration, as shown in Figure 3. This loading had an available excess reactivity, with all control rods fully inserted, of 239 lh. The worth of core material as opposed to radial blanket material was measured at the radial edge of the core. The half-core, half-blanket drawers located in both halves of the assembly in positions Q-13 and O-19 were replaced by full blanket drawers. The reactivity lost was 525 lh. Thus, the 239-lh supercriticality would be equivalent to 0.46 drawer in each half, and the just-critical loading would be 31.54 drawers in each half, equivalent to a critical volume of 34.5 liters.

Figure 3. Reference Critical Loading with RAPSODIE Engineering Core

Calibration of control rods with the engineering core was done differently than with the physics core. The reactivity effect of a given motion of one control rod was successively counterbalanced by motion of two other rods, thus calibrating the last two in terms of the first. A single period measurement for the first rod thus gives a calibration in terms of inhours. This method was found to be more precise and required less time than the method used with the physics core, i.e., determining rod worth versus position by withdrawing increments and obtaining subcritical counting rates. For the blanket-loaded rods at the core radial edge, a worth of 20 Ih/in. for the rod in 1-R-19* (see Figure 3) and 18 Ih/in. for the rod in 2-R-13 was obtained for the first 3 in. of travel. A core-loaded rod at the edge (1-N-14) gave 62 Ih/in. These values represent an increase of about 12% over corresponding calibrations in the physics core, presumably due to the decreased core size and increased ratio of plutonium to uranium.

For the critical experiment with the engineering core, the radial and axial blankets were the same as in the physics core experiments. Both had a composition approximating that of the RAPSODIE radial blanket. In the RAPSODIE design, there are steel-sodium axial end gaps (transition regions between core and blanket) just above and below the core, and axial blankets beyond the gaps. Before introducing the end gaps, the axial blankets in the critical experiment were first replaced with the proper engineering axial blanket composition. Details of the replacement and the reactivity effects are given in Table I.

Table I. Worths of Axial Blankets in RAPSODIE
Engineering Mockup

A. Compositions of Blanket Types

<u>Axial Blanket Type</u>	<u>Composition (g/cc)</u>			
	<u>Depleted U</u>	<u>Steel</u>	<u>Al</u>	<u>Mo</u>
1. Physics Fine	8.56	1.10	0.34	1.04
2. Physics Medium	11.79	0.71	0.31	
3. Engineering Axial	6.69	1.49	0.38	1.04
4. Engineering End Gaps		0.71	1.24	

B. Substitutions and Worths

Half No.	Substitution				Worth (Ih)
	Before		After		
	Axial Zone	Type	Axial Zone	Type	
1	8 to 12 in.	1	8 to 29 in.	3	-40
	12 to 25 in.	2	29 to 31 in.	4	
2	6 to 10 in.	1	6 to 19 in.	3	-30
	10 to 23 in.	2	19 to 24 in.	4	

*The first numeral indicates which half of the machine is involved. Thus, 1-R-19 indicates position R-19 in Half No. 1.

Next, the engineering gaps were introduced behind the core in each half, replacing some of the engineering axial blanket. These substitutions involved several steps with a total replacement of about $1\frac{1}{2}\%$ Δk . Furthermore, with the increased neutron leakage in the axial regions because of the sodium-steel gaps, the worths of the control rods changed somewhat, which required recalibration. The data have not yet been completely analyzed, but approximate results are as follows:

For the "lower" end gap (Half No. 2), replacement of axial blanket with sodium-steel gap for 3 in. behind the core (6 to 9 in.) gave a 300-lh loss. In Half No. 1, replacing blanket with "upper" gap for 6 in. (8 to 14 in.) behind the core resulted in a 400-lh loss. For both halves, the gap compo-

sition was the same, 0.48 g/cc sodium (48%) and 3.0 g/cc steel (38%), introduced behind the core in 32 drawers. All reactivity results are based on the conversion from period to inhours calculated for the physics core and are subject to modification when the engineering composition is calculated.

During the month, a few foil irradiations were run in conjunction with development of a technique for measuring the ratio of U^{238} captures to U^{235} fissions.

2. ZPR-VI

Measurements have been made with Assembly No. 2 of ZPR-VI. Assembly No. 2 is approximately a 650-liter carbide core, whose cross section is shown in Figure 4. Each drawer in the core contains two

$\frac{1}{16}$ -in. columns of enriched uranium, three $\frac{1}{8}$ -in. columns of graphite, four $\frac{1}{8}$ -in. columns of depleted uranium, and four $\frac{1}{4}$ -in. columns of stainless steel-canned sodium. The atomic densities and volume fractions of the materials used in the core are given in Table II.

The critical mass was 545.3 kg of U^{235} . The core height is approximately 91.4 cm, with a 30-cm-thick axial and radial high-density uranium blanket.

Control rods were calibrated by period, rod drop, and digital "rod travel" methods. Rod calibration curves, obtained by measuring the variation of the flux when the rod is withdrawn, were analyzed by means of the

Figure 4. Cross Section of Assembly No. 2 of ZPR-VI, Showing Position of Control Rods and Composition of the Blanket

usual reactor kinetics equation. Calibration curves obtained by the digital method at different rates of control rod withdrawal and different power levels agree well with those obtained by period measurements, after allowance has been made for the 0.5- μ sec deadtime of the counting system. No effects of detector location were observed.

Table II. Composition of Assembly No. 2 of ZPR-VI

	Core			Blanket		
	Atom Density, (atoms/cm ³) $\times 10^{-24}$	Average Density, g/cm ³	YOM* Volume Fraction	Atom Density, (atoms/cm ³) $\times 10^{-24}$	Average Density, g/cm ³	YOM* Volume Fraction
Stainless Steel**	0.01460	1.354	0.1724	0.006344	0.5515	0.0749
Sodium	0.007908	0.30186	0.3595	-	-	-
Carbon	0.01290	0.2573	0.1540	-	-	-
U ²³⁸	0.009850	3.892	0.2052	0.03993	15.777	0.8318
U ²³⁵	0.0022846	0.8913	0.04759	8.104×10^{-5}	0.03162	0.001688

*Yiftah-Okrent-Moldauer Cross Section.

**Assumed identical with iron.

Table III shows the total rod worths obtained by these methods, using the calculated values of $\beta_{\text{eff}} = 0.0073$ and 451 lh/% Δk . Minor changes of rod worths are expected in a final evaluation, when delayed-neutron parameters are recalculated on the basis of the measured U²³⁸/U²³⁵ fission ratio.

Table III. Total Worths of Some Control Rods

Control Rod No.	Number of Fuel Columns	Period Measurement, lh	Digital Method, lh	Rod Drop, lh
5	3	100.6 ± 0.8	99.5 ± 0.15	101.5 ± 3
6	2			80 ± 3
8	2	67.5 ± 0.5	67 ± 1	

Rod No. 5 was located at a position equidistant from the center of the core, as was Rod No. 8. However, Rod No. 5 contained three columns with $\frac{1}{16}$ in. of fuel, instead of two, in order to increase its worth.

Fission ratios of several isotopes have been determined with a pair of thin-walled stainless steel flow counters and also with a set of solid-state detectors. The measurements with the flow counters were made at the center of the assembly, while those taken with the solid-state detectors were made about 4 in. (radially) from the center. The measurements were made first with sodium-filled cans throughout the core region, then in a central voided region (~16 in. in diameter and 16 in. long) in which the sodium cans were replaced by empty cans. Ratios of atomic fission cross sections obtained in these experiments are listed in Table IV. The experimental fission ratios are considerably lower than those derived from calculated spectra, showing that the neutron spectrum is softer than expected.

The observed fractional changes of the fission ratios caused by removing the sodium from the central part, however, are in good agreement with the calculated values.

Table IV. Central Fission Ratios

Isotope Ratio	Flow Counter				Solid-state Counter			
	Sodium-filled Region σ_f/σ_f^{25}	Void Region, σ_f/σ_f^{25}	Ratio Void/Sodium		Sodium-filled Region σ_f/σ_f^{25}	Void Region, σ_f/σ_f^{25}	Ratio Void/Sodium	
			Exp	Calc			Exp	Calc
U^{233}/U^{235}	1.520 ± 0.015	1.526 ± 0.015	1.004 ± 0.008	1.006	1.45 ± 0.04	1.51 ± 0.04	1.041 ± 0.008	1.006
U^{234}/U^{235}	0.225 ± 0.003	0.250 ± 0.003	1.106 ± 0.008	1.103	0.233 ± 0.007	0.250 ± 0.007	1.073 ± 0.011	1.103
U^{236}/U^{235}	0.074 ± 0.004	0.082 ± 0.004	1.110 ± 0.015	1.091	-	0.087 ± 0.004	-	1.091
U^{238}/U^{235}	0.0346 ± 0.0005	0.0376 ± 0.0005	1.087 ± 0.008	1.075	0.0344 ± 0.0010	0.0375 ± 0.0011	1.090 ± 0.015	1.075
Np^{237}/U^{235}	0.174 *	0.188 *	1.081 ± 0.015	1.101	0.174 ± 0.006	0.187 ± 0.005	1.075 ± 0.011	1.101
Pu^{239}/U^{235}		1.062 ± 0.020			1.09 ± 0.006	1.12 ± 0.04	1.027 ± 0.008	1.023
Pu^{240}/U^{235}		0.268 ± 0.0005						
Am^{241}/U^{235}		0.110 *						

*No accurate isotopic analysis is yet available; the listed data are based on an assumed 100% isotopic purity of the Np^{237} and Am^{241} samples.

A pair of enriched and depleted uranium foils was closely attached to the sensitive area of the flow counters and irradiated for a radiochemical analysis of U^{235} and U^{238} fissions and U^{238} captures. The radiochemical fission rates are listed in Table V and are now in satisfactory agreement with those obtained directly by the fission chambers.

Table V. Radiochemical Fission Rates Compared with Fission Chamber Data

	Sodium-filled Region	Voided Region
U^{235} fission rate, * fissions/min/g U^{235}		
Radiochemical	1.279×10^7	1.109×10^7
Fission chamber	1.238×10^7	1.085×10^7
$\sigma_f^{28}/\sigma_f^{25}$		
Radiochemical	0.0345	0.0378
Fission chamber	0.0346	0.0376
$\sigma_c^{28}/\sigma_f^{25}$		
Radiochemical	-	0.0991

*Normalized to a reading of 1×10^{-8} amp on ionization channel.

The prompt-neutron decay constant, α , at delayed critical has been determined by the Rossi- α technique:

$$\alpha = \beta_{\text{eff}}/\ell = (3.50 \pm 0.10) \times 10^4 \text{ sec}^{-1}.$$

Hence, the prompt-neutron lifetime,

$$\ell = (20.9 \pm 0.6) \times 10^{-8} \text{ sec},$$

is obtained on the basis of the calculated delayed-neutron fraction $\beta_{\text{eff}} = 0.0073$.

3. ZPR-IX

ZPR-IX achieved initial criticality on February 10, 1964, with an essentially rectangular parallelepiped geometry, 47 cm on a side and 51 cm high, having a clean critical mass of 290.5 kg. In this geometry, the Rossi alpha (β/ℓ) was measured to be $7.78 \times 10^4/\text{sec}$. A power calibration was then made by means of an analysis of the statistical fluctuations of the chain reactions for Channel Nos. 3 and 4. All subsequent measurements were made after the core boundary was smoothed to approximate a circular cross section.

The core, No. 1, is a right cylinder, 62.8 cm in diameter and 51.1 cm high, whose cross section is shown in Figure 5. The clean critical mass is 279.12 kg U^{235} with a length-to-diameter (L/D) ratio of 0.814. The pre-critical estimate was 285 kg of U^{235} . The core is surrounded on all sides by a 30-cm aluminum reflector. Both the matrix and the fuel drawers are of aluminum. A core drawer contains two $\frac{1}{8}$ -in. columns of 93% enriched U^{235} and fourteen $\frac{1}{8}$ -in. columns of depleted uranium (U^{238}) with 0.2% U^{235} .

Figure 5

Cross Section of Assembly No. 1 on ZPR-IX Machine Showing Location of Control Rods and Composition of the Reflector

Control rods were calibrated under two different conditions. In the first, one of the rods was calibrated against a second rod in a different part of the opposite half of the core in order to minimize the interaction between them. In the second measurement, a previously calibrated rod was recalibrated against its image rod in the opposite half in order to

evaluate the effect of a void on rod calibration. The results showed that the differential worth decreased by as much as 0.004% ($\Delta k/k$)/cm as the rod approached its full-in position and its image rod approached its full-out position. All measurements were made by the rising-period method. Total worth measurements made by the negative ramp input method gave an integral worth of 0.42% $\Delta k/k$ and 0.34% $\Delta k/k$ for Rod Nos. 1 and 8, respectively.

A number of central reactivity worths were determined for 2 x 2 x 1-in. samples. Measurements have been arbitrarily made relative to a void and are reported below. Canned samples were sealed in Type 304 stainless steel jackets, and the quoted reactivity values have been corrected for the jackets:

Canned Void	0.27	lh/sample can
B ¹⁰	2.43	lh/g B ¹⁰
Tungsten	21.76	lh/kg tungsten
Graphite	36.5	lh/kg graphite
U ²³⁸	11.29	lh/kg U ²³⁸

The worth of U²³⁵ was also determined, both at the center of the core and at the core-reflector interface. At the center, the measured worth was +229 lh/kg U²³⁵ relative to U²³⁸, and at the edge it was +24.11 lh/kg U²³⁵ relative to the aluminum reflector material.

The main distinguishing feature of the assembly, namely, the full-density aluminum reflector, is currently being analyzed both analytically and experimentally. Of particular interest in the analysis is the effect of the aluminum resonance region and the methods of compiling resonance-region multigroup constants.

B. General Fast Reactor Fuel Development

1. Development of Jacket Materials

a. Vanadium Alloys

(i) Introduction. Jacket materials for the fuel elements of fast reactors must have suitable high-temperature properties (including strength and integrity), good thermal conductivity, and compatibility with the nuclear fuel and with liquid-metal coolants such as sodium. The jacket materials must also be amenable to fabrication into the desired shapes. Of the various potential jacketing materials that have been considered, vanadium-base alloys are among the more promising, and titanium is the element favored for use as an alloying element in the vanadium. The optimum titanium content

in the alloy is not yet known; for example, the vanadium-titanium alloy that is most inert to sodium is not likely to be the composition that is easiest to fabricate. The properties and fabricability of vanadium-titanium alloys are therefore being investigated.

Results to date indicate that compositions with 10 to 20 w/o titanium are probably most suitable, but compositions with up to at least 30 w/o are being studied. Alloys with higher percentages of titanium have been made, but their hardness is greater and they are less easy to fabricate.

(ii) Fabrication and Hardness. The vanadium-titanium alloys were made both by electron-beam (EB) melting and by arc melting (AM). In February arc-melted vanadium-base buttons were cast and fabricated

into sheet for studies of compatibility with sodium. The titanium additions were in the range of 0 to 30 w/o and were made in 5-w/o increments. Since earlier corrosion tests in sodium showed the importance of oxygen content, two sets of these alloys were made. One set was estimated to contain 300 to 500 ppm, and the other 1300 to 1500 ppm of oxygen. Figure 6 shows the hardnesses of these alloys in the annealed condition and after 50% reduction. The higher oxygen content increases the hardness of the unalloyed vanadium by at least 70 DPH points but increases the hardness of vanadium containing more than 10 w/o titanium by only 10 or 15 points.

Figure 6. Hardness of Vanadium-Titanium Alloys of Two Different Oxygen Contents

The hardness decrease in titanium alloys of high oxygen content has been previously reported by Fraser and Lund,² who have given some possible explanations. The subsequent increase in hardness with increasing titanium is ascribed to solution hardening.

Chemical analyses of specimens from two electron-beam cast ingots show that closer control over alloy homogeneity is achieved by EB melting rods composed of compacted V-Ti briquets, than by similarly melting rods prepared by welding titanium strips to preconsolidated vanadium ingots (see Progress Report for January, ANL-6840, p. 26). Accordingly, vanadium chip and titanium sponge materials have been processed in this manner into 5.8-cm-diameter alloy ingots. Three EB ingots were then

²R. W. Fraser and J. A. Lund, Effect of Titanium Additions on the Low Temperature Behavior of Vanadium, Canadian Metallurgical Quarterly, 1 (1) July-Sept 1962.

fastened together to form a consumable electrode that was arc melted to a 9.7-cm-diameter V-20 w/o Ti (TV-20) ingot. The arc-cast ingot is being inspected and will be machined into a 8.9-cm-diameter x 31.8-cm-long extrusion billet.

(iii) Elastic Modulus. The elastic modulus of the jacketing material of fast-reactor fuel elements at operating temperatures is one of the properties considered in fuel-element design. Since the elastic modulus of a material is a function of the rate at which the material transmits sound, equipment is being assembled for measuring the transit time of sound passing through potential jacketing materials at temperatures ranging from room temperature to 1100°C. A stainless steel sample will be used first for calibration.

(iv) Nondestructive Tests. The specimens to be subjected to nondestructive tests were swaged from drilled bar stock without any annealing (except of a single sample after swaging); the objective was primarily to supply tubular specimens for experimental eddy-current inspection rather than to produce tubes of maximum quality. The results, given in Table VI, do not imply that crack-free TV-20 tubing cannot be produced, but they merely indicate that the 10 w/o alloy (TV-10) is more ductile than the 20 w/o alloy. The outer diameter of the tubing was 5.72 mm and the wall thickness was 0.635 mm. The letters L and T (meaning "lead end" and "tail end," respectively) in the specimen designations refer to the part of the extrusion from which the specimen is taken, and the numbers designate the extrusion.

Table VI. Eddy-current Tests of Vanadium-Titanium Alloy Tubing

<u>Material</u>	<u>Designation</u>	<u>Length, cm</u>	<u>Results</u>
TV-10	221T	41	Numerous cracks
TV-10	221L1		No defects
TV-10	221L2		No defects
TV-20	224L	23	Numerous cracks
TV-20	224L	13	Numerous cracks
TV-20	224L	12.5	Numerous cracks
	(annealed)		

(v) Corrosion by Sodium. Specimens of vanadium-titanium alloys were exposed to flowing (15-cm/sec) sodium for 9 days at 650°C. Zirconium turnings in the sodium stream were used to getter oxygen from the system. Vanadium samples of two oxygen impurity contents and samples of niobium-10 w/o titanium were similarly exposed. The corrosion data are summarized in Table VII.

Table VII. Behavior of Vanadium and Niobium Alloys
in Sodium at 650°C

Alloy w/o	Rate of Weight Change, mg/cm ² /mo	Calculated Rate of Metal Reaction ^c	
		cm/mo x 10 ⁴	mils/mo
V ^a (SR)	+1.55	8.21	0.32
V ^a (RX)	+1.64	8.68	0.35
V ^b (SR)	+1.89	10.0	0.38
V ^b (RX)	+1.86	9.90	0.38
V-10 Ti (SR)	+1.05	5.68	0.22
V-10 Ti (RX)	+1.11	6.00	0.22
V-20 Ti (SR)	+0.254	1.44	0.06
V-20 Ti (RX)	+0.321	1.79	0.06
Nb-10 Ti (SR)	-10.2	-	-
Nb-10 Ti (RX)	-4.3	-	-

^a1070 ppm oxygen in vanadium before test.

^b200 ppm oxygen in vanadium before test.

(SR) - Stress relieved. (RX) - Recrystallized.

^cBased on the following simplifications: a) All oxygen is used in film formation. This assumption is most valid for V-20 w/o Ti and least valid for unalloyed vanadium. b) Calculated weighted average densities of the alloys: V-10 w/o Ti, 5.9 g/cm³; V-20 w/o Ti, 5.7 g/cm³. c) Film assumed to be VO.

Duplicate sodium samples gave 40 and 70 ppm (± 30 ppm) oxygen, respectively. This result is surprising (and questionable) since an oxygen concentration of about 200 ppm had been observed for the previous run. The lowest previously observed oxygen content was about 120 ppm, and no deliberate changes have been made in loop operation. Additional sodium samples from the frozen loop will be analyzed for oxygen.

The observed weight gains for the vanadium alloys are in marked contrast to previously reported behavior (see Progress Report for January 1964, ANL-6840, p. 27). This behavior could, conceivably, be the result of the apparent lower oxygen content. If so, it is of considerable theoretical as well as practical importance. It must be emphasized, however, that the reported oxygen concentration is tentative and subject to confirmation. (If the oxygen concentration, should, in fact, prove to be considerably higher, the explanation for the change in behavior must be sought in metal quality.)

The effect of titanium on the corrosion behavior of vanadium-titanium alloys is apparent. Within the range studied, the initial oxygen content has no effect on the corrosion behavior of unalloyed vanadium.

Microhardness traverses have been taken of all the samples. There was no disturbed layer discernable on the V-20 w/o Ti samples. The V-10 w/o Ti showed a disturbed layer approximately 100 μ thick, with a surface hardness of 700 DPH (100-g load) and a bulk metal hardness 160 DPH (equal to that of the vacuum-annealed control sample). The unalloyed vanadium (initial oxygen concentration of 1070 ppm) showed a shallow hardness gradient of 50 DPH from the edge to center (250 DPH surface hardness - 200 DPH bulk metal hardness, 375 μ from the surface). The center hardness of the exposed sample was 50 DPH higher than the vacuum-annealed control, indicating that oxygen had penetrated the entire sample thickness.

Titanium appears to decrease the rate of diffusion of oxygen into the metal. The relation between decreased oxygen diffusion rate and improved corrosion behavior is not understood at this time.

b. Niobium Alloys

(i) Corrosion in Sodium. Niobium-10 w/o titanium alloys were tested in flowing sodium as described in the previous section. The niobium alloys lost weight and had nonadherent coatings characteristic of niobium alloys tested in sodium contaminated with oxygen. The addition of titanium may have considerably reduced the corrosion rate as compared with pure niobium and other niobium alloys previously tested. However, the lower rates could also have been the result of the possibly lower oxygen concentration. In any case, the behavior of this material was not adequate for use as a practical fuel-cladding material.

2. Irradiation of Experimental and Prototype Fuels

a. Irradiation of Fast Reactor Fuels. In the development of fast reactors the irradiation behavior of potential and prototype fuels and fuel elements is an important factor to be determined. Three specimens, CP-33, CP-34 and CP-35 (see Progress Report for January 1964, ANL-6840, p. 25), were irradiated and examined by neutron radiography.

The uranium-plutonium-fissium fuel alloy specimens in Capsule CP-33 contained plutonium concentrations between 15 w/o and 10 w/o. The cladding material is Nb-1 w/o Zr alloy having wall thicknesses of 0.38, 0.51, or 0.63 mm. The fuel had been irradiated at a time-averaged temperature of 525°C for a calculated 2.7 a/o burnup. The radiographs revealed an average fuel elongation of 4 percent. All claddings appeared intact. In three specimens the fuel had moved off the bottom plug and areas of decreased fuel density could be seen. This capsule will be returned to CP-5 for further irradiation.

Capsule CP-35 contains six fuel pins that are of the composition U-20 w/o Pu-10 w/o Fs. Some of the specimens have a 1.4-mm axial hole;

in others the annulus between the fuel and the clad had been increased from 0.15 mm to 0.31 mm and the clad was vented. One specimen was instrumented with a central thermocouple. The fuel had been irradiated at maximum central temperatures of 680°C to a calculated burnup of 1.2 a/o. The neutron radiographs showed that the axial void was closing in the mid-portion of the fuel with an average 4 percent decrease in length. The specimens with the increased annular clearance had elongated an average of 9 percent. This capsule will be returned to CP-5 for additional irradiation.

Capsule CP-34, for which thermocouple readings had indicated a cladding failure, was neutron radiographed for confirmation of the failures and then destructively examined. The capsule contained three specimens clad in Type 304 stainless steel lined with tungsten and three specimens clad in Nb-1 w/o Zr. Three fuel compositions were used: U-20 w/o Pu-10 w/o Fs, U-15 w/o Pu-10 w/o Fz, and U-10 w/o Pu-10 w/o Fz. The post-irradiation examination showed that the stainless steel claddings had failed where the fuel penetrated defects in the tungsten barrier and made contact with the steel. The specimen fueled with U-10 w/o Pu-10 w/o Fz and clad with Nb-1 w/o Zr was intact. The other two specimens clad with Nb-1 w/o Zr alloy but containing the higher plutonium concentrations had failed. A detailed metallographic examination is in progress to study the nature of these failures.

b. Development and Fabrication of Experimental EBR-II-type Irradiation Specimens. Twenty-five fuel pins of U-15 w/o Pu-10 w/o Fz (7.7 w/o U²³⁵) alloy were loaded into Nb-1 w/o Zr, vanadium, and Nb-4 w/o V jackets. The fuel rods were closure-welded and surveyed for alpha contamination. Surface contamination was within acceptable limits. All closure welds were sound as determined by helium mass spectrometer leak tests.

Two methods were tested to calculate the quantity of sodium required: (1) volumetric measurement by immersion of fuel pin, and (2) volumetric estimation from micrometer measurement. Seventeen of the fuel rods were bonded by soaking at 500°C for 2 hr, then impacting for 1000 cycles.

Sodium levels were determined by radiography. Of the seventeen rods tested, seven had sodium levels out of the specified range. All of the fuel rods that contained fuel pins, whose volume was determined by liquid immersion, showed sodium levels within the specified range of 8.89 mm plus 3.81 mm or minus 2.54 mm. The pins whose volume was calculated from micrometer measurement of diameter and length yielded fuel rods with sodium levels out of specification.

3. Development of Carbide Fuels

For fast breeder reactors solid-solution uranium-plutonium monocarbide fuels have the advantage over solid metallic fuels of being usable

at higher temperatures and over oxide fuels of having superior thermal conductivity. A program is therefore under way for the development of carbide fuels.

a. Production of PuC and (U·Pu)C and Fabrication of Carbide Fuel Element Specimens. The production of solid-solution $(U_{0.8}Pu_{0.2})C$ by a diffusion process is presently being investigated. Powders of the individual carbides are used as starting materials. During the month of February a mixture of -80 mesh (U.S. Standard) powders was fired at 2000°C for a total of 40 min at 10-min intervals. After each interval, the furnace was cooled, the fused cake removed, sampled, and reground to the initial particle size.

The two photomicrographs, Figures 7 and 8, show the structure of the fired cake after 10 and 40 min. PuC is very much in evidence after

Figure 7

UC-20 w/o PuC after 10 min at 2000°C. 200x; atmosphere etched.

Figure 8

UC-20 w/o PuC after 40 min at 2000°C. 200x; atmosphere etched.

10 min, but has diffused into the UC with time until only a few intergranular areas show PuC after 40 min. The third and fourth firings gave a marked increase in bulk density.

One objective of these experiments is to produce a solid-solution carbide suitable for fabrication into fuel specimens by vibratory compaction. This requires dense particles some of which are as large as +12 U.S. sieve size. The next series of experiments will use UC of the +12 sieve size for starting material and -80 sieve size for the PuC component.

Four vibratory-compacted specimens containing physically mixed UC-20 w/o PuC were fabricated. The cladding was 0.144 in. (3.66-mm)-ID Nb-1 w/o Zr. The fuel was loaded to a depth of 2 in. (5.08 cm). The empty cladding was wrapped with aluminum foil before introduction into the glove-box. The samples were compacted and the top end plug inserted. The foil was removed immediately before welding. The maximum activity after welding was 400 dpm. No loose activity was found. Based upon this limited experience, the use of a protective foil appears to reduce surface contamination to an acceptable level without introducing the welding problems which accompany the use of such protective coatings as collodion.

b. Transient Irradiations. Mixed UC-20 w/o PuC and PuC powders, vibratory compacted in either Type 304 stainless steel or Nb-1 w/o Zr tubing, have been irradiated in TREAT to determine the performance and stability of these potential fuel systems. Two TREAT capsules, which will provide calibration data for all future tests, have been returned from Idaho. The specimens are being analyzed for burnup to obtain a value of the energy release in the specimen as a function of integrated reactor power.

4. Radiator-type Fuel Elements

Five powder specimens of U-15 w/o Pu-10 w/o Fz have been successfully sodium bonded by melting of a measured amount of sodium over the previously evacuated compact and pressurizing the system. During this month's activity, eleven vibratory-compacted specimens were bonded by the method previously reported (see Progress Report for January 1964, ANL-6840, p. 22). With a sodium level not greater than 1.5 mm ($\frac{1}{16}$ in.) over the compacted powder level considered acceptable, three specimens were accepted.

An attempt was made to recover the eight rejected specimens by centrifuging at 300°C and 2100 rpm. Five of these eight, which showed high sodium levels indicative of poor bonding, were accepted after centrifuging. The remaining three specimens, which have resisted "injection" bonding and centrifuging, were made up of -325 U.S. sieve size powders. The sodium-bonded specimens are being cleaned prior to closure welding.

With the work on spherical shapes completed, the activity for this month was directed toward the compaction of irregular shapes. Malleable steel grit, as obtained from a producer of blast cleaning materials was classified into fifteen size fractions from -10 +12 U.S. sieve size to -325 mesh. The pycnometer density of a sample from each size fraction was determined. The average density was 7.28 g/cc, with a standard deviation for each various size fraction being 0.05 g/cc.

Over ninety separate data points have been obtained during the vibratory compaction of a single-component system of irregular shaped grit. The packing efficiency of the irregular shape versus particle size was compared against the model developed for spherical shapes. It appears that a similar packing system exists; however, in the case of the irregularly shaped particles, the influence of particle size is extended beyond the boundary condition; that is, beyond a ratio of container diameter to particle diameter of 10, where the influence of the boundary condition becomes negligible, the packing density of irregular shapes decreases with decreasing particle size. This phenomenon is being studied while binary compaction studies are being carried on.

5. Zero-power Reactor Fuels

a. U-25 w/o Pu-1.5 w/o Fe Fuel Plates. Theories used in the design of new reactors are generally tested to some extent in zero-power reactors (ZPR), a device used to determine the effects on reactivity and other physical characteristics of changing the number, type, and arrangement of fuel elements. ZPR fuel plates, like those of other new reactors, need to be developed and fabricated. Different types of fuel plates may be required for the physical studies.

Three precision casting melts of ZPR-III-type fuel plates, in graphite or copper molds, were made as indicated below:

<u>Quantity</u>	<u>Thickness, mm</u>	<u>Length, mm</u>	<u>Width, mm</u>	<u>Mold Material</u>
3	2.18	50.8	50.8	Graphite
6	2.18	76.2	50.8	Graphite
3	4.50	50.8	50.8	Graphite
4	4.50	76.2	50.8	Graphite
3	8.99	50.8	50.8	Copper

Mold temperatures of 300°C and 550°C were used. Metallographic examination of the resulting structure is under way. The as-cast alloy appears to be quite brittle and to resist oxidation.

Hot rolling was unsuccessful on the bare cast plates exposed to the helium atmosphere of the glovebox. A network of cracks appeared after

the third roll pass (5% total reduction). Subsequently, two plates sealed in mild steel jackets were rolled to 50 percent reduction. Evaluation of the rolled material is incomplete; however, the rolled alloy is brittle and difficult to handle, as evidenced by its fragmentation upon clamping and handling during dejacketing.

6. Nondestructive Tests of Fast Reactor Tubing

Type 304 stainless steel tubing, 9.53-mm OD by 0.51-mm wall, is being inspected in lengths of 1.5 m by pulsed-field electromagnetic test equipment and by gamma radiography. The electromagnetic scanning in this case is longitudinal. Twelve passes are made on each tube while the tube is rotated at one axial revolution per minute, so that the area being inspected on any pass lies along a shallow pitch spiral. The test equipment is adjusted to reject any tube containing a localized reduction in effective wall thickness of over 10%. Of the 30 tubes tested this month, 8 were free of defects and 11 contained usable lengths (approximately 1 m) of defect-free material. Although evaluation of the defective areas is not complete, apparently most of the defects are radial cracks starting from the inner surface. The radiographic tests revealed voids in four tubes.

C. General Fast Reactor Fuel Reprocessing Development

1. Melt Refining of Uranium-Plutonium-Fissium Fuels

The melt refining of uranium-plutonium-fissium alloy beneath a molten salt layer is being investigated as an alternative process, for which the advantages of higher yields and lowered operating temperatures have been demonstrated in small-scale experiments. It is anticipated that the rare earth elements in the fissium will be preferentially oxidized into the salt phase and thereby separated from uranium and plutonium. In current studies, the degree of plutonium vaporization and of plutonium transfer into the flux during processing is being measured. The amounts of plutonium evaporated and extracted into the flux with magnesium chloride oxidant absent (see Progress Report for January 1964, ANL-6840, p. 30) were 0.05% and 0.09% of the plutonium charged.

In the most recent chloride flux-melt refining run, the flux consisted of 81 w/o BaCl_2 -19 w/o CaCl_2 to which was added 110% of the theoretical amount of magnesium chloride oxidant. Uranium-plutonium-fissium alloy containing 0.335 w/o cerium was used. Process conditions were identical with those in the preceding run - the charge was held at 1150°C for one hour under argon. Approximately $10^{-3}\%$ of the plutonium vaporized, and 0.45% was extracted into the flux; the cerium-plutonium separation factor was 130. This high removal of cerium and relatively good retention of plutonium in the metal phase indicate that use of an oxidant (MgCl_2) with a halide flux is a promising melt-refining technique.

2. Skull Oxidation

The skull which remains in a zirconia crucible after melt refining of an EBR-II uranium-fissium charge is oxidized to convert the skull into a powder which can be readily poured from the crucible. To determine the residual uranium content of zirconia crucibles after removal of oxidized skulls, six crucibles which had been used for melt refining of uranium-fissium-0.65% cerium charges were individually pulverized and sampled. X-ray spectrochemical analysis of the six samples gave an average uranium content for the crucibles corresponding to 2.5% of the uranium originally in the skull, or 0.18% of that originally charged to melt refining. Similar values are expected when the oxidation step is performed in the EBR-II Fuel Cycle Facility.

3. Skull Reclamation Process

The skull reclamation process which is currently under development for the recovery of uranium from skull material is based on the use of molten metals and salts as solvent media (see Progress Report for August 1963, ANL-6780, p. 22). A second continuous skull-reclamation run has been performed up to the final retorting step in large-scale (1.5 kg of uranium) integrated equipment (see Progress Report for January 1964, ANL-6840, p. 30 for previous run data). All components of the system performed well in the present run.

Since backflushing of argon through the transfer tube (the usual procedure for keeping the transfer tube open) can cause splashing of solutions to the cooler upper wall of the crucible where some of the splashed material may no longer be available in further process steps, a different procedure was evaluated in the present run: The metal was allowed to freeze as a plug in the transfer tube during processing, and the plug was to be melted by heating the transfer tube when a transfer was to be made. This freeze-plug technique was successful only for the noble metal-extraction step. Heating of the transfer tube following the precipitation steps and product dissolution step did not melt the plug. Therefore, backflushing with argon (at a lower rate than was previously employed) was necessary prior to transfer of waste and product solutions from the latter steps. The appropriate technique (freeze-plug or backflushing) will be used in process steps of subsequent skull reclamation runs.

4. Eddy Current Induction Probe

An eddy current induction probe for the remote determination of liquid metal levels (see Progress Report for November 1963, ANL-6808, p. 25) has been tested for possible use in skull reclamation and other pyrometallurgical processes. The metals employed in preliminary tests of the usefulness of the probe were cadmium, cadmium-8 w/o magnesium, and

magnesium. The probe (enclosed in a stationary well) was lowered below the surface of the molten metal, and a change in the output signal of about 2 mV/in. of immersion was observed for each metal. When used with the two cadmium solutions, the probe was insensitive to the first $\frac{3}{4}$ in. of depth. The reason for this is being further investigated.

Temperature affected the output signal. With the 4-in. probe completely below the surface level of a pool of cadmium-8 w/o magnesium, the effect of temperature was 0.002 mV/°C. With no melt in the furnace, the effect of temperature on the output signal was 0.00765 mV/°C. This indicates that, with a probe of the present design, a calibration must be performed to determine temperature corrections over the entire range of immersions.

D. Sodium Coolant Chemistry

Heretofore, the two common methods of sodium oxide analyses in flowing sodium, namely, plugging valve and distillation methods, have often produced divergent results. Efforts to correlate the results of the two methods have led to a thorough study of the limitations inherent of each. The study of the distillation method has revealed that, in addition to meniscus trouble (see Progress Report for January 1964, ANL-6840, p. 32) in withdrawing sodium samples for oxide distillation analyses, it has been found that a minute film of distilled sodium has been sticking to the outside of the sampling cup. When this sampling cup was immersed in water to dissolve the remaining sodium oxide, the sodium film on the outside of the cup also dissolved, thus producing extremely high results. This trouble has been corrected by the simple expediency of wiping the exterior surface of the cup with moist tissue before transferring the sodium oxide residue out of the cup into the beaker for titration. Test results show a greatly improved accuracy in a six-sample run withdrawn from a loop.

E. EBR-II

1. Reactor Plant

a. Primary Sodium Pumps. Assembly and final inspection of sodium pump No. 2 in the presence of the manufacturer's representative were completed. The NaK-filled pressure transmitter for the pump was delivered and has been calibrated before being assembled to the pump. After replacement of the pressure transmitter, the pump was installed in the primary tank on February 25.

b. Fuel Gripper. During the first half of the month, the gripper shaft assembly, which had been binding, was thoroughly cleaned of sodium and sodium oxide by immersion in alcohol and water. The gripper jaw, bellows, and actuating shaft assembly were then dried by heating and evacuation. The bellows was helium leak-tested successfully. Operation of the

gripper jaws and the sensing rod checked out satisfactorily. The entire gripper mechanism was found to be in good condition without signs of significant wear.

After removal of the gripper mechanism from the rotating plug, gauges were passed through the gripper guide sleeve and the baffles in the reactor vessel cover, indicating that clearances are normal. The cause of sticking experienced at the end of the wet critical experiments was probably due to accumulation of sodium oxide in the annular clearance between the gripper shaft and the guide sleeve in the reactor vessel cover during the many periods of fuel handling with repeated raising and lowering of the gripper and cover. Flow of primary sodium during normal pump operation should sweep out or dissolve any oxide tending to accumulate in the gripper sleeve. Therefore, a recurrence of sticking due to oxide accumulation is considered improbable, particularly in view of the corrective action described below.

The gripper standpipe was removed and modified to provide for a split aluminum bronze insert at the top and an "O" ring groove on the flange face for sealing with the packing gland flange. Previously, the standpipe had a close clearance fit with the gripper shaft.

The outer gripper tube was modified by drilling three $\frac{3}{4}$ -in.-diameter holes about 23 in. from its bottom to provide argon gas balancing during vertical gripper shaft movement. Four $\frac{1}{2}$ -in.-diameter holes were drilled just above the lower bearing in the tube to promote mixing of the bulk sodium with the sodium in and above the bearing.

The gripper shaft assembly was installed in the primary tank on February 11, and reassembly of the components external to the primary tank was completed a few days later.

c. Control Rod Drives. The No. 8 control rod drive shaft assembly, which had been removed to permit installation of the oscillator, was thoroughly cleaned of sodium and sodium oxide and dried. The bellows in this assembly was successfully helium leak-tested after cleaning. Operation of the control rod gripper jaws and sensing rod was checked and found to be satisfactory. The assembly was put in storage.

d. Oscillator. The installation of the oscillator drive, control system, and associated instrumentation was completed. The oscillator rod and thimble are being reworked at Argonne, Ill.

e. Seal Heater Control System. Checkout and adjustment of the system continued.

f. Sodium-purification System. Installation of shielding for the primary sodium-purification piping and surge tank is essentially complete.

g. Cover-lifting Device. Early in the month, the lead screws and nuts for the lifting drive were replaced. Those originally installed were known to be worn from previous service and also were subjected to an excessive loading late in January during preparations for checking the bore in the guide sleeve and baffles of the reactor vessel cover. Inspection of the nuts revealed that some of the threads were cracked.

During an initial attempt to lift the reactor vessel cover, the newly installed lead screws and nuts became badly galled. The old lead screws, which are in reasonably good condition, were then reinstalled with bearing-bronze nuts. Initial operation of the cover lifting drive was successful, using the bronze nuts and the original screws.

h. Fuel-transfer Port. Maintenance and modification of the fuel-transfer port were completed. Three badly worn "O" rings in the port were replaced. A manual release which can override the electrical interlock was added for operation of the port.

i. Fuel-element-rupture-detection Loop (Delayed-neutron Monitor). Piping for this loop was completed except for the tie-in at the E-3 nozzle of the primary tank.

j. Inert Gas System for Primary-tank Annulus. The inert gas system for the primary-tank annulus was completed and satisfactorily tested near the end of the month. This system is intended to maintain an atmosphere of nitrogen gas in the annular space between the inner and outer walls of the primary tank.

k. Reactor Building Containment. Pressure relief check valves have been installed in the personnel and emergency air locks to permit any excess air pressure to bleed into the reactor building. Gauges were also installed to indicate pressure inside the air locks.

The suspect exhaust, purge air supply, and outside air-intake isolation valves have been tested. The total measured leak rate is satisfactory with regard to criteria established for reactor building containment. The purge exhaust isolation valve was found to have a high leak rate. The cause of the leak is being investigated.

2. Sodium Boiler Plant

a. Secondary Yard Piping. During the last part of January, the caps were cut off the 12-in. secondary sodium lines from the intermediate heat exchanger. The return bend in the yard piping was also cut off in preparation for fit-up and welding of the 12-in. spool pieces which will connect the intermediate heat exchanger with the secondary sodium system.

Fit-up and welding of the 12-in. spool pieces and of three 2-in. lines (two secondary system vent lines and the primary system fill line) were completed. Radiography of the completed 12-in. and 2-in. welds disclosed apparent defects in several welds of the 12-in. spool piece, presumably due to "lack of penetration" or "lack of tie-in." Of the total of six 2-in. welds, four were acceptable while two showed defects requiring rework.

The field welds on the 12-in. spool pieces were visually inspected at the pipe interior through holes drilled in the pipe. After this inspection and trial rework of some weld defect areas, four field welds were rejected. The spool pieces were cut out, and replacement spool pieces were fit up in preparation for welding.

The lack of weld penetration may have resulted from excessive argon gas pressure in the system. Control of pressure in the large secondary system was difficult. A further apparent complication was convective flow of argon gas through the secondary piping and the intermediate heat exchanger, which was at a temperature of about 260°F. Aspiration of air through openings in the pipe wall and contamination of the argon might have occurred. Some areas of the welds on the pipe interior appeared to be oxidized, lending credence to the latter theory.

To maintain close control of argon purge rate and pressure during the welding of the new spool pieces, balloons will be inflated in the 12-in. piping, thereby isolating the spool pieces and the weld zones from the remainder of the system. The balloons will be placed in the pipe through $\frac{3}{4}$ -in.-diameter holes drilled in the pipe wall. At each end of a spool piece, near the weld zone, two diametrically opposed holes of $\frac{3}{4}$ -in. diameter are being provided for purge gas inlet and venting and for visual inspection of the interior weld during welding.

b. Secondary Cold Trap. While the fit-up and welding of the yard piping was proceeding, the 2-in. piping associated with the replacement secondary cold trap was fit and welded. Radiography of six welds made in this piping disclosed a number of defects in the welds and the pipe-to-fitting joints. Because of the piping configuration, fit-up had been very difficult, accounting for some of the faulty joint preparation. This piping will be removed entirely and the piping configuration changed to permit better fit-up.

c. Other Work. The cooling system for the secondary sodium recirculating pumps was converted from Dowtherm to plant cooling water service to provide better cooling of the pumps.

Calibration of the NaK-filled differential pressure transducer for the main secondary flowmeter was completed. The transducer had been removed from the system since it could not be calibrated in place. Preparations are proceeding for reinstallation of this transducer, equipped with valves for future in-place calibration.

3. Power Plant

All power plant systems were in shutdown status during the month. The steam generator was maintained in wet lay-up completely filled with water containing morpholine and sodium sulfite. Weekly samples of water from the steam drum indicated a pH of about 9.7 and a sulfite ion concentration of about 90 ppm.

Nonreturn valves supplied by the turbine manufacturer were installed in the low-pressure and high-pressure turbine extraction lines in addition to the nonreturn valves initially installed. These have been added, in accordance with the turbine manufacturer's recommendation, because of the large amount of energy available downstream in these extraction lines. The control air lines to the operating cylinders for these valves were partially installed. The air lines will be completed when the actuators are installed.

The turbine manufacturer recommends installation of actuators similar to those for the first two nonreturn valves. The actuators will be inside the oil tank and will be operated by hydraulic oil pressure to the turbine stop valve. Closing of the nonreturn valves would therefore be automatic upon turbine trip-out. These new actuators are not yet on hand.

The plant cooling water system has been taken out of service and drained for installation of an acid injection line to the tower basin and installation of a relief valve between the cooling water return and suction lines. The cooling tower basin was flushed thoroughly to remove accumulated sediment and debris. The function of the relief valve is to prevent overpressurization of the condenser cooling water side by the discharge head of the reactor auxiliaries cooling water pump or the plant cooling water pump if all valves on the return water header at the cooling tower were to be inadvertently closed.

Valves were installed in the plant cooling water and reactor auxiliaries cooling water lines to permit future operation of crucial equipment around the plant with raw water cooling if the normal cooling water supply must be interrupted for system maintenance or because of equipment failure.

The cooling water system was refilled and returned to normal service.

Two improvements were made to the turbine-generator lube oil system. A transfer pump was installed, capable of moving all the oil from the dump tank to the main oil tank in less than half an hour. A lube oil filtration system was also installed, capable of recirculating oil from the main oil tank through the filters at a rate of 3 gpm. The transfer pump and oil filter system were tested; performance was satisfactory.

Design of a warm-up system for the feedwater and blowdown lines was completed and procurement of materials was initiated.

4. Training

The preparation of supervisors for approach to power operation and reactor operator systems training for technicians continued.

Seven shift supervisors and five foremen participated in a series of sessions on a review of control systems, power plant operation, determination of reactor transfer function, wet critical experimental results, and the approach to power experimental program.

5. Fuel Cycle Facility

Following the initial testing of equipment in the Argon Cell, air was admitted and personnel entered to perform minor repairs and modifications, as well as to remove mercury which had been blown out of the blister seal (located in the ceiling) during a purge of the blister (see Progress Report for August 1963, ANL-6780, p. 18). A short circuit in the wiring for the inoperative No. 2 crane bridge (see Progress Report for January 1964, ANL-6840, p. 36) was located in the termination of a mineral-insulated cable at the inner end of the bridge. Tests of a hoist motor starting under load indicated relatively high voltage transients between terminals could occur intermittently and cause breakdowns of the type experienced here. The M-1 cables on both crane bridges have been replaced by No. 12AA asbestos-insulated wire contained in electrical conduit. Also, slip clutches were installed on hoist drives for the crane and roof blister, manipulator hoist drums were modified, and new manipulator bridge drive couplings were inserted.

Hardened bevel gears were installed in transfer lock drives, and shutter drives were modified to permit their removal and replacement without use of the manipulator articulated mechanical arm attachment.

Additional work in the Argon Cell included modification of clips on the removable light fixtures to avoid sticking and contact failure. Tempered-glass cover plates were installed on the inner face of all shielding windows to protect the lead glass from impact and possible damage by internal discharge following gamma irradiation.

One of the two Argon Cell circulating fans was removed for examination. Since some wear was observed on one bearing, new bearings were installed; the fan and motor combination was rebalanced before replacement. Inspection of the purification-system compressor showed the condition of the rear bearing to be good after operating more than 1,800 hr. The Deoxo catalyst was removed and replaced with fresh catalyst. All planned Argon Cell work was completed and the cell refilled with argon.

The drive and cutter portions of a decanning unit were installed in a glovebox in the Lab and Service Building. In this location, the effects on

decanning operations of variation in oxygen and water content, cutting and drive speed, and tool setting were investigated. An investigation of materials used for cutting tools has been initiated in Illinois.

Nine additional melt refining runs, which were carried out remotely, have been completed in the two melt refining furnaces in the Argon Cell. These runs were carried out primarily to prepare uranium-fissium alloy for use in the injection casting furnace. The liquation times for these runs ranged from $\frac{1}{2}$ to 1 hr. In all runs, unirradiated, uranium-fissium chopped pins were used. In two of the runs, sodium-coated pins were used and in one of these two runs, the heel from an injection casting crucible was melt refined with the pins. The poured yields varied from 85.6 to 96.4%; the low yield of 85.6% is attributed to the large amount of slag which was introduced with the heel from the injection casting crucible.

Seven melts were made in the injection casting furnaces during the last month. The gas flow to the number one furnace actuator was adjusted and mold heaters and mold thermocouples were tested and operated.

The tantalum-tungsten protection tube for the immersion thermocouple was damaged in handling. It worked satisfactorily for eight melts.

The pin-processing unit at Window 5 (Mod-A manipulator service) processed more than 100 pins successfully. Calibration of the weight and length modules was accomplished. Initial calibration of the volume, density, and porosity modules is under way.

Initial attempts at calibration of the sodium-level and sodium-void signals of the bond-test machine indicated that the associated electronic system was not adequate. Various modifications are being studied.

Various other equipment has been adjusted and made operable.

Following satisfactory operational tests in the EBR-II mockup area at Argonne, the plant-scale equipment for the oxidation of the residue (skull) remaining in the crucible after melt refining has been shipped to Idaho for installation in the Argon Cell. Six skulls from the melt refining of unirradiated uranium-fissium alloy (cerium content, 0.6 w/o) were used in the testing of the oxidation furnace and the process control system. The skulls which ranged in weight from 591 to 902 g were processed in oxidation times of 379 to 582 min. Testing of the skull oxide dumper was also carried out. After each skull oxidation run, the oxide was transferred by means of the dumper from the melt refining crucible to a storage container. The performance of the dumper was considered to be satisfactory; the maximum loss of the oxide powder was 7 g.

The assembly machine (see Figure 9) and assembly-machine auxiliaries have arrived from Argonne, Illinois.

Figure 9. Fuel-element-assembly Machine

Figure 10 shows a tool designed for handling No. 6 food-pack cans with the operating manipulator of the Fuel Cycle Facility. This is a two-stage tong device operated by the double-hook manipulator grip. When the grip is closed part way, the tongs grip the can. The tongs are spring loaded to prevent crushing the can but hold with sufficient firmness that the loaded can may be transported in the vertical position. If the grips are closed (see Figure 11), the tongs will rotate allowing the contents of the can to be poured.

Figure 10. Food-pack Can Held in Transfer Position by Tong of Operating Manipulator

Figure 11. Can Held in Pouring Position

The forked tip of the fuel rod is easily bent in remote handling and the bonding operations. A tip-straightening press which coined the tip between dies was developed during EBR-II Core I fabrication. This tool was modified for master-slave manipulator operation as shown in Figure 12.

6. Examination of Control Rod

One control element, which had been "scrammed" several times and subjected to excessive acceleration, was removed from EBR-II and shipped to the Argonne, Illinois site. The element was cleaned, disassembled, and will be inspected as follows:

- (1) Radiograph;
- (2) Cold eddy current test;
- (3) Hot eddy current test;
- (4) Remelt of sodium;
- (5) Cold eddy current test.

The first two steps have been completed. The X-ray negatives show that nearly all the rods have shrinkage voids under the restrainer. The presence of the voids has caused all the sodium levels to be high. The average level above the fuel pin was found to be 0.78 in. (1.97 cm). Acceptable sodium level tolerances were 0.65 ± 0.10 in. (1.65 ± 0.254 cm) above the fuel pin.

Figure 12. EBR-II Fuel Element Tip Straightener

Traces for the cold eddy current test carried out to determine the sodium bond integrity are being compared with standard traces. The results of that comparison are not yet available.

F. FARET

1. General

The United Engineer and Construction Company of Philadelphia has been selected to provide general construction management services for FARET. Working relationships between the Laboratory, the Commission, the Bechtel Corporation, and the United Engineer and Construction Company are being defined.

The feasibility of reducing rock excavation requirements and minimizing the use of concrete piers under reactor building foundations was investigated and it was concluded that any possible gains were insufficient to warrant changing from the foundation plan described in the Title I report.

Title II design by the Laboratory, in coordination with the AEC, has covered the following areas:

a. The contents of the first construction package, which covers site development and excavation, have been determined. Drawings and specifications are being developed.

b. Piping and instrumentation diagrams for the sodium-purification system, inert gas system, and certain plant-process systems have been received for preliminary information and review.

c. Representatives of potential suppliers of major sodium-system and inert gas-system equipment were informed of equipment-operating requirements and equipment specifications.

d. Special emphasis has been placed on problems associated with the cell argon system and considerations of containment.

e. The state of the art of sodium-purification system design and practice has been reviewed. Detailed design criteria for the FARET purification system have been prepared and transmitted to the architect.

f. Axial, multigroup, source flux calculations have been completed by the Laboratory and transmitted to Bechtel for use in shielding design.

g. A review has been completed of preliminary control room and control panel layouts.

h. An alternative proposal for cooling the interior of the cell has been evaluated. This design provides for use of propane-cooled refrigeration

units instead of the water-cooled heat exchangers described in the Title I report. The basic proposal was generally acceptable. However, the Laboratory has requested reconsideration of the refrigerant (ammonia was suggested in lieu of propane). Maintenance of all cell-cooling units must be feasible regardless of cell inaccessibility considerations.

i. A radiation zoning plan for the reactor building has been established.

j. Extensive information concerning Laboratory-designed equipment, plant-operating requirements, and general design criteria required for the development of the Title II design have been transmitted to the architect.

2. Stress Analysis

The preliminary design of the FARET grid support consists of a circular, perforated, simply supported plate reinforced by a simple beam from below. The plate is subjected to a uniformly distributed transverse load and to temperature gradients along its thickness and the radial direction. This is illustrated in Figure 13. In order to give a combined maximum deflection of 0.015 in., the following loading conditions were assumed for analysis:

- uniformly distributed transverse load = 117 lb/in.^2 ;
- linear temperature gradient across the thickness = 50°F ;
- linear temperature gradient along the radius = 50°F ;
- ambient temperature = 1000°F .

Figure 13. Sectional View of FARET Grid

A description of the procedure used in obtaining the stresses involved consistent deformations of one plate and the supporting beam.

The bending stress in the outer fiber of the beam is given by the relation

$$\sigma = \frac{3Px \left[\frac{2}{\ell}(m-p)x + p \right]}{b \left[\frac{2}{\ell}(m-p)x + p \right]^3 - (b-t) \left[\frac{2}{\ell}(n-q)x + q \right]^3}.$$

In this equation P denotes the total load transmitted to the beam by the grid, ℓ is the length of the beam, and x the distance from one end of the beam to the point where stresses are being considered. The other quantities are defined in Figure 13.

The shearing stress in the beam at any cross section is given by

$$\tau = \frac{3}{4} \frac{ba^2 - (b-t)h^2}{ba^3 - (b-t)h^3},$$

where a and h (the variable distances between the upper and lower portions of the beam) become equal to p and q at the supports and to m and n at midspan, respectively. The resultant maximum stresses in the beam, as related to its physical dimensions (in in.), is shown in Table VIII.

Table VIII. Maximum Stresses in Supporting Beam

<u>b</u>	<u>t</u>	<u>m</u>	<u>n</u>	<u>p</u>	<u>q</u>	<u>τ max, psi</u>	<u>σ max, psi</u>
20	2.5	14	10	4	0	9,080	4,080
22	2.5	12	8	6	2	6,050	4,660

3. Safety Analysis

The second draft of the FARET Preliminary Safety Analysis Report (PSAR) has been completed and reviewed by the Laboratory's Reactor Safety Committee. The Committee's suggestions are being evaluated and revisions related thereto are being developed.

A code for the 7094 Digital Computer has been developed by Bechtel for investigation of the performance of the reactor coolant system. Results obtained for a variety of system conditions will be included in the Preliminary Safety Analysis Report.

4. Cell

A computer program has been written for the LGP-30 to obtain some information on the dynamic behavior of the FARET cell atmosphere. The program provides for varying several parameters the absolute values of which are not known at the present time, such as inleakage rate, purification rate, and purity of the gas returned from the purification system. Other variables that can be handled are rate of introduction of makeup argon,

outleakage rate to the vessel around control rod drive seals, and shutdown of the purification system. To date, the program has been used only to obtain changes in the oxygen concentration in the cell.

5. Component Development

a. Investigation of Heat Transfer Cements. The merits of encasing resistance-type sodium pipe heaters with a heat transfer cement has been investigated. Three characteristics were studied:

- (1) reduction of the heater surface temperature when encased in cement;
- (2) capability of the cement to provide a more uniform pipe surface temperature;
- (3) physical characteristics of cement after exposure to 1200°F for long periods of time. Two commercially available cements were tested.

An 8-ft section of 8-in.-diameter, Type 304 stainless steel, Schedule 40 pipe and a $\frac{5}{16}$ -in.-diameter resistance heater were assembled. Thermocouples, placed as shown in Figure 14, recorded the surface temperature of the heater, the air temperature within the pipe, and the internal surface temperature of the pipe. The $\frac{5}{16}$ -in. resistance heater was covered with an angle iron. Approximately one-half of the pipe and heater assembly was covered with 3 in. of insulation, the other half with 5 in. of insulation.

The section of pipe with 3 in. of insulation was used to determine the difference in heat transfer characteristics of the two cements that were tested. Any difference was experimentally negligible.

The section of pipe with 5 in. of insulation was used to determine the effectiveness of the cement to reduce heater surface temperatures and diametrical pipe temperature differences. The surface temperature of the heater was found to be 123°F lower in the heater section encased in cement as compared with the heater section covered only by the angle iron. Diametrical temperature profiles of the internal surface of the pipe are shown in Figure 15. The internal pipe surface temperature in the cement-encased heater section ranged from 870°F to 787°F for a total diametrical ΔT of 83°F. In the noncement-encased section, the internal surface temperature of the pipe ranged from 908°F to 823°F for a total diametrical ΔT of 85°F.

Two samples of one product exposed to 1200°F became brittle and spalled badly within 750 hr. The other product showed no adverse effects after exposure for 1525 hr.

TEMPERATURE °F OF CAL ROD SURFACE AND AIR IN PIPE

TCX#1	TCX#2	TCX#3	TCX#4	TCX#5	TCX#6
832	1036	1159	716	670	714

NOTE: TEMPERATURES AN AVERAGE OF NINE READINGS
POWER INPUT ON THE HEATER = 131 WATTS/FT AVG

- TCX#1 AIR TEMPERATURE AT PIPE CENTER
- TCX#2 SURFACE TEMPERATURE OF CAL ROD UNDER CEMENT (5 INCHES OF INSULATION)
- TCX#3 SURFACE TEMPERATURE OF CAL ROD UNDER ANGLE IRON (5 INCHES OF INSULATION)
- TCX#4 SURFACE TEMPERATURE OF CAL ROD UNDER CEMENT (3 INCHES OF INSULATION)
- TCX#5 SURFACE TEMPERATURE OF CAL ROD EXPOSED TO AIR
- TCX#6 SURFACE TEMPERATURE OF CAL ROD UNDER CEMENT (3 INCHES OF INSULATION)

Figure 14. Pipe-Thermocouple Layout

Figure 15. Internal Temperature Profile on Pipe

b. Oxygen Meter. The electrochemical oxygen meter cell (see Progress Report for August 1963, ANL-6780, p. 27), which uses a solid zirconia electrolyte for continuously measuring oxygen concentration in sodium, has been assembled and connected to the test loop. All major leaks in the sodium zone of the loop were eliminated. Leaks in the gas zone, around the small spark plug-type liquid-level indicators were sealed with high-temperature epoxy resin. The loop has been filled, and circulation and cold trapping has been started to establish equilibrium in the system.

An earlier test indicated a temperature of $\sim 100^{\circ}\text{F}$ at the region of the silicone "O"-ring seal, which separates the gas phase of the two cells, when the loop temperature is $\sim 700^{\circ}\text{F}$. Temperature checks at the seal, now actually installed in the system, give slightly higher readings, but these are well within the temperature range recommended for silicone rubber. The temperatures measured were:

<u>Loop Sodium</u>	<u>Silicone Rubber Seal</u>
250°F	110°F
495-505°F	145°F

The seal is backed up with asbestos packing and a packing gland to protect the seal from sodium vapor.

6. Fuel Development

a. CP-5 Irradiation Loop. A mockup of the CP-5 irradiation loop to test both its mechanical performance and the heat transducer characteristics has been completed. Installation and removal of the fuel capsule into the thimble can be readily accomplished before and after heat cycling.

To test the heat transducer (see Progress Report for January 1964, ANL-6840, p. 45), various heat fluxes up to approximately 36 W/cm were applied by means of electric current. For specimens 6 in. long, and an observed ΔT of 45°F , the heat flux will be 30.2 ± 1.3 W/cm. The temperature differences for one thermocouple located at the horizontal centerline and for one $6\frac{15}{16}$ in. down (see Figure 16 of ANL-6840) are shown in Figure 16. The expected error was of the order of ± 20 W over the whole range. Although the thermocouples used were not of good quality, they were sufficient to establish the feasibility of the transducer operation in the range up to approximately 30 W/cm. Instability observed above this level may be attributed to the quality of the thermocouples. Thermocouples of higher quality will be installed and the test repeated. It is also planned to extend the range of use of the transducers to heat fluxes up to 300 W/cm.

The safety, installation, and operational aspects of the in-pile experiment have been reviewed by the CP-5 reactor management. The apparatus has been approved for installation in CP-5 at the VT-27 thimble location. Shop fabrication has been started.

b. Heat Transfer Bench Tests. The thermal-cycle bench tests of uranium carbide pellets for the FARET Doppler experiment have been continued (see Progress Report for December 1963, ANL-6810, p. 20). Two additional tests (UC #2 and UC #3) have been run with two 2.8-in. lengths of carbide pellets. As previously, the pellets were fabricated at the Laboratory by cold pressing, vacuum sintering, and electric-discharge machining to pellet densities of about 80 to 83 percent of theoretical. Further

work will be required to increase the fuel densities to approximately 95 percent of theoretical. Fissures were present in some pellets, but to a lesser extent than in the previous samples.

Figure 16. Temperature vs. Heat Input for Transducer Test

The cladding for the pellet test section described previously has been shortened to accommodate four 2.8-in. lengths of pellets. Lava pellets are used to fill spaces not occupied by carbide pellets. The pellets are now centered by dimples in the cladding, and the 2.8-in. lengths of pellets are each supported by rings resting on cladding dimples. (The original test section and details of the cladding dimples and pellet supports are illustrated in the Progress Report for December 1962, ANL-6672, p. 23, and in the Progress Report for April 1963, ANL-6717, p. 28, concept No. 7, respectively.) The test section is enclosed (see Figure 17) in a plastic bag, filled with argon in order to exclude oxygen from the test section if a leak should occur. This would prevent a rapid carbide-oxygen reaction at high operating temperature.

(i) Test UC #2. The appearance of the pellets before testing is shown in Figure 18. The pellets were thermally cycled nine times between heat fluxes which had been estimated to be about 60 W/cm and 10 W/cm, with a 0.5-hr period. These conditions would cause roughly three times greater stresses than for the 20-W/cm maximum flux and one-hour period used during the first UC test. However, when the data were analyzed, it was observed that the maximum clad temperature was only 640°C

Figure 17. Thermal-cycling Test Section Encased in Plastic Bag

Figure 18. Pellets before Test UC #2

versus 580°C for the first test, indicating that the maximum heat flux during the second test was not really much higher than during the first test. In addition, after nine cycles, the heater burned out (concluding the test) outside the carbide test region. This served to confirm the suspicion that higher temperatures, together with higher resistivities and heat fluxes, had occurred outside the carbide test region.

After the test, only two pellets were found to be cracked. In Figure 19, pellet I-1 (pellet I-5 in Figure 18) was cracked during the loading operation, and pellet II-5 (pellet II-2 in Figure 18) may have been cracked by thermal stress, but the geometry of the pellet was not affected. The dull appearance of the pellets after test is due to surface oxidation. It is probable that either impure argon cover gas or absorbed air in the pellets caused this.

Figure 19. Pellets after Test UC #2

(ii) Test UC #3. The appearance of the pellets before testing is shown in Figure 20. Only those pellets labeled 1 through 4 were new, the other pellets having been used during the second test above. To insure that the heat fluxes would be known relatively accurately, the tantalum heater length was reduced to the length of the carbide test region, 15 cm. To keep the resistance of the heater high, its OD was also reduced from $\frac{1}{16}$ in. to $\frac{1}{32}$ in. In addition, the heater was insulated electrically from the carbide pellets, which exhibited high electrical conductivity, by a ceramic tube. The pellets were cycled ten times between heat fluxes of about 63 W/cm and 13 W/cm, with a 0.5-hr period. Then the power was raised until the heater burned out, at about 78 W/cm. Test conditions are given in Table IX. As shown in Figure 21, the pellets suffered no apparent damage during the test.

For the next UC thermal-cycle tests, it is planned to use pellets which are drilled to permit installation of thermocouples inside the

carbide. It should then be possible to obtain additional data concerning full temperature changes which may be achieved by changing the gas pressure in the gap between fuel and cladding.

Figure 20. Pellets before Test UC #3

Table IX. Conditions for Test UC #3

Heat Flux (W/cm)*	13	63	78
Cladding Temperature (°C)*	480	710	745
Carbide OD Temperature (°C)**	730	1270	1360
Carbide ID Temperature (°C)**	750	1360	1470

*Measured.

**Calculated.

Figure 21. Pellets after Test UC #3

7. Sodium Flow in Fuel Assembly

A facility for testing FARET fuel subassemblies in the high-temperature sodium environment has been designed. Investigations will include thermal and flow studies, alignment of fuel, and in-core instrumentation. In addition, considerations for fuel handling and remote handling of components have been included. Thought has been given to future adaptation of the facility for duplicating cell conditions over the test vessel.

The facility, shown schematically in Figure 22, consists of a pump, pressure vessel, expansion tank, dump tank, heat exchanger, and connecting piping. The loop will provide FARET reactor conditions, being capable of operating at 1200°F with a maximum flow rate of 800 gpm of sodium.

Figure 22. Facility for Testing FARET Fuel Subassemblies, Schematic

The design of the pressure vessel and fuel subassembly support structure is similar to that of the actual FARET facility. The fuel subassembly support structure will be removable and has been designed to accommodate a 19-element core.

An interim pump capable of delivering 230 gpm sodium at a 110-ft head has been obtained from LASL. Specifications have been completed for the purchase of an 800-gpm, 320-ft-head sodium pump.

III. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. Thick-target Neutron Yields for 6-BeV Protons

In an effort to evaluate the possibility of using very high-energy accelerators as a neutron source for time-of-flight spectroscopy, 6-BeV protons from the Bevatron were used to bombard a thick target located in the center of a large water tank. The resulting neutron flux distribution in the water was measured by foil activation. The proton beam intensity was measured by the $\text{Al}^{27}(\text{p}, 3\text{pn})\text{Na}^{24}$ reaction. The results for targets of 10 cm in diameter by 15 cm long were:

	<u>Neutrons/Proton</u>
Uranium	113 $\pm$ 25
Lead	27 $\pm$ 5
Aluminum	1.7 $\pm$ 0.4
D ₂ O	1.7 $\pm$ 0.4

The results in neutrons/proton incident for targets 10 cm in diameter for various target lengths were

	<u>Target Length</u>		
	<u>5 cm</u>	<u>10 cm</u>	<u>15 cm</u>
Uranium	34	74	113
Lead	8	18	27
Aluminum	-	-	1.7
D ₂ O	-	-	1.7
Background Yield	0	0.02	0

Error averaged $\approx \pm 20\%$.

2. Mass-Energy Relations in U²³⁸ Fission

A measurement of the mass and energy distribution of 1.5-MeV neutron fission of U²³⁸ has been made. About 28,000 fissions were recorded. Measurements of U²³⁵ thermal fission were made at the same time with the same equipment. Preliminary results are given in Table X and Figures 23 and 24.

Table X. Preliminary Results for Fission Constants
Obtained from Experiments with 1.5-MeV
Neutrons Measured from Zero Degrees of
Neutron Beam with the Solid-state Detectors

<u>Measurement/Material:</u>	<u>U²³⁵</u>	<u>U²³⁸</u>
<u>Neutron Energy:</u>	<u>Thermal</u>	<u>1.5 MeV</u>
Average Mass of Light Peak: $\overline{M}_L$, amu	96.1	98.8
Variance of Light Peak: σ_L^2 , amu ²	36.9	38.9
Average Mass of Heavy Peak: $\overline{M}_H$, amu	139.7	139.5
Variance of Heavy Peak: σ_H^2 , amu ²	36.7	36.8
Average Total Kinetic Energy: $\overline{E}$, MeV	165.6	167.6
Variance of Total Kinetic Energy: $\sigma^2(E)$, (MeV) ²	99.3	101.4

Figure 23. Average Total Kinetic Energy vs. Fragment Mass. Measurements made with 1.5-MeV neutrons and solid-state detectors at zero degrees from neutron path.

Figure 24. Normalized Yield vs. Fragment Mass. Measurements made with 1.5 MeV neutrons and solid-state detectors at zero degrees from neutron path.

3. Angular Distributions of the $\text{Li}^7(p,n)\text{Be}^7$ Reaction near the Threshold

Preliminary measurements of the angular distributions of the ground-state neutron group for the $\text{Li}^7(p,n)\text{Be}^7$ reaction have been obtained. A B^{10} glass scintillator was used in conjunction with time-of-flight techniques to make the measurements. The data are being analyzed to obtain differential cross sections.

4. Elastic and Inelastic Scattering of Fast Neutrons from Tl, Hg, and Au

The elastic and the inelastic scattering of 300-1500-keV neutrons from Tl, Hg, and Au has been studied by time-of-flight techniques. The respective differential cross sections were measured at incident-neutron intervals of $\lesssim 50$ keV. The inelastic excitation of a number of states was observed including; (a) 205, 280, 620, 680, 920, and 1080-keV levels in Tl; (b) 160, 210, 350, 440, 650, and 980-keV levels in Hg; and (c) 77, 270, 540, 830, 940, and 1220-keV levels in Au. A number of the observed states have not been previously reported.

The measured elastic scattering was compared with optical model calculations by means of the spherical potential of Moldauer.³ The Hauser-Feshbach method was used to obtain inelastic excitation function for comparison with experiment.

³P. A. Moldauer, Optical Model of Low Energy Neutron Interactions with Spherical Nuclei, Nuclear Phys. 47, 65-92 (1963).

5. High-conversion Critical Experiment

A sequence of measurements were made on the factors affecting conversion ratios in densely loaded fuel zones of the High Conversion (Hi-C) Critical Experiment. Among the advantages of such cores are their relatively high conversion ratios, which conserve fissionable material and also reduce long-term reactivity transients in power reactors. Measurements were made on critical mass, reaction rates among the various constituents, buckling, and other factors which are helpful in understanding the physics of these cores.

Measurements were started in January with an assembly having a stainless steel-clad central fuel zone on a 1.27-cm triangular lattice core. Upon completion of the measurements that core was replaced by a second core loaded on a 1.17-cm triangular pitch with 4,900 aluminum-clad elements. Uranium oxide enriched to 3 w/o was used as fuel in these experiments. The fuel was loaded as pellets, 0.94 cm in diameter, into either aluminum or stainless steel tubes which have a diameter of 1.06 cm. The fuel length in each tube was 122 cm.

The ratios of epi- to subcadmium captures in U^{238} (ρ_C^{28}) and the fissions in U^{235} (ρ_f^{25}) were calculated from cadmium ratio (C) measurements in the 1.27-cm pitch stainless steel-clad fuel zone. The dysprosium substitution technique gave a value of $C_{28} = 1.185$, based on three measurements, with an estimated standard deviation of 0.003. The average of eight direct measurements (bare capture/cadmium-covered capture) of C_{28} was 1.24 ± 0.01 (see Progress Report for October 1963, ANL-6801, p. 39). Extrapolation to zero thickness of cadmium and the effects of other corrections reduced this value to $C_{28} = 1.20 \pm 0.01$. The two techniques failed to agree within the range of applicable correction terms. It has been concluded that the substitution method is preferable provided that the dysprosium activation and U^{238} capture follow expected behavior. The advantages included better reproducibility and lessened dependence on cadmium thickness. The following corrected measurements are reported:

$$\begin{array}{ll} \rho_C^{28} = 5.4 \pm 0.1 & \rho_f^{25} = 0.36 \pm 0.01 \\ C_{25} = 3.75 \pm 0.1 \text{ (fuel location)} & \zeta_{25} = 1.11 \pm 0.02 \text{ (thermal} \\ C_{25} = 4.14 \pm 0.1 \text{ (moderator location)} & \text{disadvantage factor)} \end{array}$$

The second core with the fuel on a 1.17-cm triangular pitch had a theoretically clean critical cylindrical loading, which required 5,091 fuel elements containing 115.7 kg of U^{235} and 3,741 kg of U^{238} . The actual loading was reduced to 4,900 elements, to provide room for 20 water channels. Finger-rod guide tubes were placed in these locations. The excess reactivity was 0.3%; the volume ratio of $H_2O:UO_2$ was 0.44; and the atomic ratio of $H:U^{238}$ was 1.33. Insertion of 20 fuel elements at the guide tube locations would reduce reactivity by 0.58% because of water displacement and local changes in the $H:U^{238}$ ratio. A gain of 0.0016% per fuel element was measured on adding fuel at the periphery.

This core has the tightest uniform loading in which the available fuel supply produces criticality. Measurements being made in this core will complete a sequence of measurements in fuel zones ranging upward from an H:U²³⁸ atomic ratio of unity, for which results were reported earlier. Other future work will include remeasurement of a few values obtained in other cores in which the dysprosium substitution method was not applied.

The need for the development of a pile oscillator method for study of capture-to-fission ratios in fuel of critical experiments is also under study. Because of the hardened neutron-energy spectrum in these cores it is not accurately known what fraction of neutrons are captured without fission in U²³⁵.

6. Gain Stabilization of Analog Amplifiers in the Nanosecond Regime

Many nuclear and reactor physics experiments require the amplification of analog signals (fast-rising voltage or current pulses) from a nuclear detector at bandwidths of the order of 200 Mc/sec, either to maintain precise timing or to keep pulse widths short and thus avoid pileup and overloading. At such large bandwidths, it is difficult to stabilize the amplifier gain and, to some extent, the discriminator threshold, in such a way as to make these units insensitive to changes and fluctuations in circuit element characteristics; in fact, the gain in most wide-band amplifiers (especially those of the chain or distributed type) is not stabilized at all.

All devices and systems which have been used to improve the stability of nuclear analog information acquisition and processing equipment (detectors, photomultipliers, pulse amplifiers, and pulse-height and -shape discriminators of various types) make use of feedback in some way. The best and simplest application of feedback is internal degenerative feedback. However, the benefit from this method deteriorates when the signal transit time through the feedback circuit elements becomes comparable with the inverse of the upper half-power frequency. Beyond this point, external feedback may be used. External feedback denotes a method where a suitable sample of the pulse-height spectrum, or an injected test signal, is compared with a standard and the error signals derived from this comparison are used to adjust the gain. This stratagem is labeled "superregulation." This system requires time to redress the performance of the equipment, and therefore it passes a certain amount of false information, depending on the frequency and extent of the "natural" excursions of the analog equipment and the frequency of inspections. The use of a superregulator does not, therefore, allow one to dispense with careful temperature and supply-voltage regulation if very precise results are required.

The superregulation scheme considered here was designed for a system having a relatively stable converter of nuclear particle energy to current pulses (fission chamber) but which can tolerate unstable pulse amplification in the amplifier and somewhat uncertain threshold stability in the discriminator. The discriminator is used in the external feedback loop with a simple digital logic circuit to pulse channel gain. A block diagram of a count channel superregulator bench test device is shown in Fig. 25. In addition to the wide-band amplifiers, discriminator, and scaler being tested, the system includes a highly stable low-voltage supply, a double pulser of charged cable type, a flipflop, three coincidence circuits, a bias control circuit, and assorted wide-band adder and splitter units, delays, and attenuators.

Figure 25. Block Diagram of the Bench Device Used to Test Gain Stability in a Count Channel Designed with Superregulator

To determine the gain stability of a given amplifier or group of amplifiers, the clock drive is usually set to 1 pulse/sec, and the unit may then be left to run for several hours, or even days. The power for the amplifiers may be supplied directly from the ac lines or through ac regulators. Some typical plots of gain variation which were obtained in this way are shown in Figure 26, which, incidentally, illustrates the improvement in gain stability which results from the use of ac regulation (this behavior was observed with several different types of chain amplifiers). For the sake of comparison, a run is also shown in which the pulser output was fed directly to the discriminator through an appropriate delay.

Figure 26

Plot of Results of a Count Channel with the Superregulator Having a Relatively Stable Detector and Amplifiers Having No Internal Stabilization. In Curve A, the amplifiers were powered directly from the ac line ; in Curve B, the amplifiers were powered through ac regulators and show improved performance; and in Curve C, the output of the pulser was fed directly to the discriminator.

The preliminary tests carried out thus far with the particular superregulator design demonstrates that it improves channel gain stability, at least when the detector (as in the case of a fission chamber or solid-state device) is relatively stable and the amplifiers have no internal stabilization. The degree of improvement necessarily depends on the type, as well as the operating history, of the particular amplifiers used.

When the gain adjustment steps are small, and the test pulses are sufficiently short, around 10 nsec, to be readily accepted by the discriminator, count rates of the order of $10^6/\text{sec}$ are well within the capability of this type of regulator.

B. Theoretical Reactor Physics

1. Studies of the Doppler Effect

Extensive studies have been made in order to include the contribution of interference interactions of neighboring resonances as well as resonances of the fertile isotope on the Doppler broadening of fissile isotopes. Calculations have been carried out for both Pu^{239} and U^{235} at various temperatures and potential scattering cross sections.

The effect of interference interactions was found to be strongly dependent on energy, enrichment ratio, and the type of fissile isotope under consideration. The percentage of reduction in the Doppler broadening from the corresponding value by the isolated resonance approximation versus the ratio of atomic densities is plotted in Figures 27 and 28. The dotted curves in Figure 28 show the reverse effect of Pu^{239} on the Doppler broadening of U^{238} . The results seem to indicate that U^{235} will exhibit negative Doppler effect in all atomic ratios of practical interest so long as it is homogeneously mixed with the fertile isotope.

Figure 27

Percentage of Reduction in Doppler Broadening
vs. the Atomic Ratio of U^{238} to Pu^{239}

Figure 28
Percentage of Reduction in Doppler
Broadening vs. the Atomic Ratio of
 U^{238} to U^{235}

2. Statistical Theory of Nuclear Collision Cross Sections

A formalism has been developed for a statistical treatment of the energy variations of nuclear scattering and reaction cross sections. A statistical collision matrix, $\underline{U}^S$, is defined which has the form of an energy-independent direct transition matrix plus a fixed simple resonance pole expansion, the matrix residues of which are products of complex channel width amplitudes. By direct comparison with the Wigner-Eisenbud and Kapur-Peierls collision matrices it is found that under widely applicable conditions the statistical collision matrix may be used to calculate averages of observables over energy intervals containing many resonances and many total widths. The problem of determining the statistical properties of the parameters of $\underline{U}^S$ has been solved for several special cases by relating it to the statistics of R-matrix parameters.

By means of these methods, averages and mean square fluctuations of total and reaction cross sections have been calculated under general conditions admitting direct and compound processes, and arbitrary average values of the total widths, Γ , and the resonance spacings, D . The results are expressed in terms of the direct reaction matrix elements and the statistical properties of resonance parameters appropriate to the energy region under consideration, and they are related to the locally applicable optical model phase shifts and transmission coefficients.

Simplifications were obtained under special assumptions, such as uncorrelated width amplitudes, small and large Γ/D , pure compound nucleus reactions, many competing open channels, and many competing direct processes. In the limit of small Γ/D , one obtains the leading terms of an expansion of the average cross section which had previously been derived from R-matrix theory directly. In the limit of large Γ/D (many competing channels but no direct reactions), the nonelastic fluctuation (or average compound nucleus) cross sections approach the Hauser-Feshbach formula. Except in this limit, corrections due to partial-width fluctuations and resonance-resonance interference are applicable. The former are sensitive to the magnitudes of direct reaction matrix elements, the latter to the correlations of resonance energies. Competing direct reactions have been shown to require reductions of the transmission coefficients. The mean-square fluctuations of cross sections are found to approach Ericson's results in the limit of large Γ/D and many competing channels, but they are in general much larger for moderate Γ/D and few channels. They are also sensitive to the details of resonance parameter statistics.

3. Total Neutron Cross Sections

A Fortran code has been written which permits the calculation of the potential, resonance, and interference cross sections for up to 50 single-level Breit-Wigner resonances with natural line shape at specified energy

single-level Breit-Wigner resonances with natural line shape at specified energy values. Orbital angular momentum values of $\ell = 0, 1$, and 2 are allowed. The code will be used to make preliminary studies of the effects of nearby resonances on the cross sections near a given resonance energy.

4. Resonance Scattering in Light Elements

Aluminum is often used to simulate sodium coolant in fast critical experiments. The relative reactivity effects of sodium relative to those of aluminum are then found by replacement substitution in various regions of the core. Sodium and aluminum differ in their dependence of the resonance scattering cross section upon energy in the neutron range of fast systems. Theoretical analyses of replacement experiments are generally carried out with multigroup cross-section parameters having group-energy increments which encompass many scattering resonances. Furthermore, the multigroup parameters used may also have been a-priori evaluated by taking into consideration the differing fine-group structure of the sodium and aluminum-containing regions. Still unaccounted, however, is the possible effect of the resonance-produced fine-structure flux spectrum of the surrounding aluminum-containing region upon the effective group cross section of the internal sodium-containing region, and vice versa.

Indicative of the possible effects of the adjacent aluminum-containing regions in modifying the effective group cross sections for transport and elastic transfer in the sodium-containing region are the values given in Table XI for an EBR-II core composition and a ZPR-III

Table XI. Ratios of Transport and Elastic Removal Cross Sections
Obtained by Aluminum Relative to Sodium Resonance-
produced Fine-flux Weighting

Energy Group ⁴	EBR-II		Assembly No. 31	
	Transport*	Elastic Removal**	Transport*	Elastic Removal**
2	1.00	0.98	1.00	0.97
3	1.00	1.035	1.00	1.06
4	1.00	0.93	1.00	0.94
5	1.01	0.82	1.01	0.81
6	1.00	0.93	1.01	0.91
7	1.00	0.93	1.01	0.90
8	1.00	1.10	0.99	1.175
9	0.99	1.48	0.98	1.61
10	1.02	0.67	1.03	0.59
11	1.00	1.98	0.95	2.02
12	0.975	0.75	0.945	0.525
13	1.00	1.04	1.00	0.99
14	0.99	0.93	0.99	0.97
15	1.07	0.17	1.25	0.19

*Ratio of total homogenized core cross section for transport.

**Ratio of light elements (sodium and stainless steel) homogenized core cross sections for elastic transfer.

⁴Yiftah, Okrent, and Moldauer, Fast Reactor Cross Sections, Pergamon Press, New York (1960).

Assembly 31 core composition. Listed are the ratios of effective coarse-group cross sections of the sodium-containing composition determined by resonance-produced fine-group fluxes of the aluminum-containing composition divided by the effective coarse-group cross sections of the sodium-containing composition determined by the resonance-produced fine-group fluxes of the sodium-containing composition. The modifying effect is seen to be greater for the elastic removal than for the transport cross sections in these cases. These results were obtained by averaging the fine-group cross sections of the ELMOE⁵ library for composition of the sodium-containing systems with the fine group fluxes. The latter were determined with the ELMOE code for the aluminum- and sodium-containing cores, respectively.

5. Romberg Integration Algorithm

The Romberg integration algorithm has been used with great success by many groups^{6,7} and appears to be among the most generally reliable quadrature methods available. It is, therefore, worth pointing out that it is not entirely foolproof, and that a significant class of integrands exists for which the extrapolated values are poorer estimates of the integral than the corresponding trapezoidal sums.

Any integral for which the odd derivatives of the integrand either vanish, or are equal at the limits, will not be improved by Romberg extrapolation. Among the common examples of such integrals are those of periodic functions over a period and those for which the derivatives vanish at both limits.

C. High-temperature Materials Development

1. Resistance to Corrosion by Lithium

Whenever higher operating temperatures can be effected in power-reactor design without undue complications, costs, or impairment of safety, a resulting increase in the efficiency of the reactor can be expected to result in cheaper power. One difficulty is that the higher operating temperatures introduce problems in cooling the reactors. Some of these problems would be solvable by the use of a high-boiling, low-density coolant, such as lithium, provided suitable corrosion-resistant containers for the coolant could be fabricated at a reasonable cost. Lithium (and some of its impurities formed by lithium-air reactions) is, however,

⁵H. Hummel and A. Rago, An Accurate Treatment of Resonance Scattering in Light Elements in Fast Reactors, Seminar on Physics of Fast and Intermediate Reactors. Vienna (1962).

⁶Henry C. Thacher, Jr., Certification of Algorithm 60, Comm. ACM 5, 168 (March 1962).

⁷Karl Heinz Buchner, Certification of Algorithm 60, Comm. ACM 5, 281 (May 1962).

corrosive to many metals at high temperatures. A program has therefore been instituted to study the resistance of materials to corrosion by lithium at elevated temperatures.

An inert atmosphere is necessary when working with lithium if the required high purity is to be maintained. The usual procedure for maintaining such an atmosphere involves the use of a glovebox, a relatively expensive piece of equipment that occupies valuable space even when not in use. For certain specialized operations, which are performed intermittently, e.g., material transfer, an alternative approach would be desirable.

Accordingly, a "glove pouch," illustrated in Figures 29 and 30, has been developed. The bag is made of vinyl sheet with two 40.6-cm (16-in.) receiving windows and a pair of 11.4-cm (4.5 in.)-ID gloveports to which are attached neoprene gloves. Figure 29 is an overall view of the facility. Figure 30 is a close-up view of a portion of the lithium melting and casting apparatus.

Macro 38430

Figure 29. Vinyl "Glovepouch" Containing an Inert Atmosphere for Work with Lithium.

The glove pouch was evacuated and purged with argon by repeated (about six times) deflation and inflation, and then sealed. Purging and lithium transfer was completed in less than 30 min. Lithium is very sensitive to even low concentrations of nitrogen and water vapor in the atmosphere, and excessive levels are indicated by the formation of a black film on the surface. The transfer was accomplished with no visible film formation.

As can be seen from Figure 29, the glove pouch can be set up at any convenient work space. It is deflated when the operation is completed and is reusable.

2. Sintering Evaluation of Uranium Sulfide

Two batches of uranium monosulfide, designated

Macro 38429

Figure 30. A Partial View of Apparatus for Melting and Casting Lithium in a Glovepouch.

54M and 57, were made by the gas-solid reaction and were homogenized before transferring. Oxygen contents of 0.21 and 0.09 percent and unit cell sizes of 5.489 and 5.490 Å were reported for 54M and 57, respectively, in the homogenized conditions.

Each of the batches were crushed to pass an 80M (U. S. Standard) sieve and were ball milled for 6 hr in CCl_4 . The resulting powders were mixed with 1% stearic acid and pressed into 0.952-cm-diameter pellets, which were vacuum sintered for 3 hr at 1650°C.

Table XII gives data on the sintered specimens. Chemical analyses were not obtained. The decrease in unit cell size with sintering probably resulted from a pickup of oxygen while the material was a fine powder.

Table XII. Density and Cell Size of Sintered US and the Effect of Homogenizing on the Cell Size

<u>Batch</u>	<u>Sintered Density, % Theoretical</u>	<u>Cell Size, Sintered, Å</u>	<u>Cell Size, Homogenized, Å</u>
54M	94.4	5.484	5.489
57	93.5	5.487	5.490

Specimens were polished and etched for metallographic examinations. Both specimens were made up of two phases, the minor phase being UOS. The UOS in the specimen of 57 was restricted to the grain boundaries and appeared to have been molten as it took on the shape of the boundaries; in the specimen of 54M the UOS was present as small, rounded discrete grains located mostly in the grain boundaries.

3. Properties of UP, US, UP-US, and ThP

The volatilization of several compositions in the UP-US system was studied. The results of firing in vacuum are presented in Table XIII. All samples showed small weight losses on firing at 1800°C, although the US sample exhibited a somewhat larger loss. Compositions fired at 2200°C showed more distinct trends of weight loss. UP exhibited the least weight loss, whereas the composition UP-90% US showed the greatest. The weight loss versus composition relationship was not linear, since the intermediate compositions showed somewhat larger losses than might be expected.

Table XIII. Volatilization of Compositions in the UP-US System

	<u>UP</u>	<u>UP-30% US</u>	<u>UP-90% US</u>	<u>US</u>
Weight Loss/hr, at 1800°C, %	0.20	0.22	0.31	0.46
Weight Loss/hr, at 2200°C, %	1.42	2.41	3.75	3.28

Thorium monophosphide powder was mixed with binder and cold pressed into pellets. The powder had a lattice constant of 5.834 Å, corresponding to a theoretical density of 8.83 g/cc. After being fired at 1800°C for 2 hr, the sintered samples had an average density of 77% of theoretical. The interior of the samples was blue, as was the original powder, but a thin outer layer was silvery. It is possible that this layer is the subphosphide of thorium ($\text{ThP}_{0.7}$), which is mentioned in the literature. The lattice constant of the fired pellets was virtually unchanged from that of the original ThP powder. Otherwise, only weak lines for ThO_2 were observed in the X-ray powder photograph of the fired sample.

D. Other Reactor Fuels and Materials Development

1. Titanium Alloys

a. Corrosion by Mercury. Despite its high density and low boiling point, mercury has the advantages as a reactor coolant that it is more easily purified than many other liquid metals, and it does not attack some metals, including titanium, as aggressively as other liquid metals do, particularly when they contain the products of reaction with air. Furthermore, there is evidence that the presence of certain additives in the mercury make it even less aggressive in its attack on titanium.

The effect of a third additive (ppm of aluminum or magnesium) to nickel-containing or zirconium-containing mercury on the corrosion behavior of titanium was investigated. The use of both metallic additives was based on the assumption that they may have some scavenging and/or wetting effects. Results of a 14-day exposure at 538°C in a static system are in

Table XIV. The data reveal that there was little effect produced by aluminum or magnesium addition to the zirconium-containing mercury. However, greatly accelerated attack on titanium was observed in the Hg-Ni-Al and Hg-Ni-Mg systems.

Table XIV. Interaction of Crystal Bar Titanium and Additives
Exposed to Liquid Mercury at 538°C (Static)

Test Conditions	Weight Change, mg/cm ² after 14 Days	Spectrochemical Analysis of Exposed Surfaces*				
		Hg	Ni	Zr	Mg	Al
Liquid Hg	-16.51	W	-	T	-	-
	-18.69	W	-	T	-	-
	-7.33	W	-	T	-	-
Liquid Hg + Zr (750 ppm)	-0.15	M	-	M	-	-
	-0.94	M	-	M	-	-
	-0.92	M	-	M	-	-
Liquid Hg + Zr (750 ppm) + Al (50 ppm)	-0.89	V-VS	-	-	-	VS
	-1.56	V-VS	-	-	-	VS
Liquid Hg + Zr (750 ppm) + Mg (50 ppm)	-2.08	M	-	S	-	-
	-1.07	M	-	S	-	-
Liquid Hg + Ni (750 ppm)	+0.04	M	M	-	-	-
	+3.92	M	M	-	-	-
Liquid Hg + Ni (750 ppm) + Al (50 ppm)	-28.56	M	W-M	-	-	W-M
	-29.87	M	W-M	-	-	W-M
Liquid Hg + Ni (750 ppm) + Mg (50 ppm)	-34.10	W	T	-	-	-
	-31.20	W	T	-	-	-

*VS - very strong, above 10%

V - strong, 10 to 1%

M - moderate, 1 to 0.1%

W - weak, 0.1 to 0.01%

T - trace, 0.01 to 0.001%

It appears that the effects of the addition of one or more metallic additives to the Ti-Hg system involve a complex mutual interaction. In general, beneficial effects of an inhibiting material are largely attributable to the formation of an insoluble protective film. If the film is not thermodynamically stable or if no film can be formed, the corrosion of the titanium will be adversely affected. Thus, nickel or zirconium addition was effective; addition of aluminum or magnesium to a Hg-Zr system was of little effect; the addition of aluminum or magnesium to a Hg-Ni system was deleterious.

2. Zirconium Alloys

a. Fabrication. In the continuation of a study of the influence of alloy microstructure on corrosion behavior, samples of steam-resistant zirconium alloys have been worked by a high-energy forming (HEF) technique. As-cast samples in ring form (similar to tubing sections) were

submitted to the Laboratory of General Atomic Division for diameter reduction by their MAGNEFORM process, a magnetic pulse technique. A diameter reduction of about 10% was achieved at room temperature (for 5-cm-OD rings of 1.5-mm wall thickness). The ring form was chosen to simulate a fuel-clad section and to demonstrate a conceivable clad application technique, as well as to obtain HEF material. The samples are being submitted to corrosion test and metallographic examination.

b. Corrosion in Superheated Steam. Zirconium alloys that are relatively corrosion resistant to steam at 650°C and 42.2 kg/cm² (600 psig) are being tested in water vapor at 650°C and 1.35 x 10⁻³ kg/cm² (1 torr absolute) in order to (a) observe the effect of steam pressure on the corrosion rate, and (b) relate the results of electrical studies made in the 1 torr range to corrosion processes at higher pressures. Samples of Zr-3 w/o Ni-0.5 w/o Fe, and Zr-1.2 w/o Fe-1.1 w/o Cu, pre-exposed 0.9 day to the high-pressure steam, were transferred to the lower-pressure environment and tested for an additional 2.7 days. Specimen weight gains were comparable with those of samples tested continuously at the higher pressure for this time. The test is being continued to obtain data over a 12-day range at the lower pressure.

Long-term exposure of Zr-4 a/o Ni and Zr-4 a/o Cu alloys (as cast) to steam at 540°C, and 42.2 kg/cm² has continued. After 385 days the Zr-Cu samples continue to gain weight at a low rate. Total weight gained was 6.2 mg/cm² (avg of 3). The rate of gain of the Zr-Ni samples was somewhat variable, totaling 9.4 mg/cm² (avg of 3) after 385 days. Thin edge cracks of the film were apparent at 13X on all samples, but there was no surface blistering. The corrosion product was adherent and colored gray to white.

3. Ferrous Alloys

a. Corrosion in Superheated Steam. The 18 percent chromium, 8 percent nickel types of austenitic stainless steels do not resist corrosion by superheated steam as well as do some of the straight-chromium stainless steels (such as Type 406) or as well as do the aluminum-modified higher-nickel alloys (such as Incoloy 800), or perhaps even the aluminum-modified 18-8 type chromium-nickel stainless steels. The effects on corrosion behavior of changing the temperature or pressure of the steam or of modifying alloy compositions are not well known in all instances. The corrosion behavior in superheated steam of the relatively corrosion-resistant ferrous alloys is therefore being investigated in detail.

Type 406 stainless steel brazed with Coast Metal 60 has been exposed to flowing (approx. 30 m/sec) steam for 56 days in a continuing test. Temperature was 650°C, pressure 42 kg/cm². The braze experienced corrosive attack, particularly in the excess braze material along the sides

of the joint. Most of this has been converted to corrosion product and is easily chipped off with a probe. The joint itself did not appear weakened, and the corrosive attack is proceeding only slowly into the braze alloy between the joined 406 plates.

Incoloy 800 modified with aluminum (approx. 4%) has now been exposed 14 days to flowing (60 m/sec) steam at the above temperature and pressure. Metal loss was about 0.4 mg/cm², a relatively low value compared with alloys tested previously.

The new dual-wall autoclave system for higher temperatures and pressures has been completed and tested. Types 304 and 406 steel have been exposed to steam at 750°C and 42 kg/cm² for one week. The metal loss values for Type 304 were only slightly higher than those obtained at 650°C. The Type 406 samples have not been defilmed as yet, but the existence of a temper film indicates that lower metal corroded values are to be expected at 750°C than at 650°C for this alloy, at least for short exposures.

4. Studies of Polarization in Sodium

Reaction layers on the surfaces of metals are known to inhibit the corrosive attacks of certain media. In some instances an imposed electric potential has been used to produce or maintain the corrosion-inhibiting properties of the film when the potentially corrosive medium is a liquid metal. A program for studying the effects of polarization in a liquid metal environment therefore offers promise of overcoming some corrosion problems. Part of such a study has involved the investigation of the current-limiting properties of films built upon zirconium alloys in oxygenated sodium.

A corrosion film formed earlier in oxygenated sodium at 650°C on Zr-3 w/o Ni-0.5 w/o Fe material was found to be a poor polarization current-limiting component in the 650°C sodium cell with a cathodic current of about 8 mA/cm² applied. A specimen of Zr-1.2 w/o Fe-1.1 w/o Cu alloy having a substantial film formed in 650°C steam was then exposed in the sodium cell at 650°C and 2.8 mA/cm² cathodic current. The current-limiting property of this film too appeared to deteriorate after a short time. On removal after 2.7 days, a small weight loss was observed, and the film seemed to have been partially transformed into a two-layer product. The underlayer was a tough, black, adherent insulator at room temperature. Further experiments are planned with this type of film including an attempt to retain the film anodically.

The cell is presently being modified to eliminate the development of internal short circuits produced by condensation of sodium vapor. In brief tests of potential insulators, Viton O-rings survived a 0.7-day test

at 185°C in sodium vapor. Teflon developed a black surface at 185°, a gray-black discoloration at 160°, and a slight gray surface at 123°C in similar tests. Slow reduction to carbon is implied, but the possibility of a limited usefulness near the melting point of sodium is indicated.

5. Nondestructive Testing

a. Ultrasonic Imaging. A sample-positioning mechanism has been completed that facilitates ultrasonic inspection of clad fuels during passage of inspection samples through the inspection area at a rate of 75 cm/min. The steady movement of inspection samples by this mechanism has been of great value in observing defect areas in these samples. As an example, a BORAX-V superheater fuel plate (a trial manufacturing plate made from depleted uranium) has been used for some time to demonstrate the ability of the imaging system to detect edge defects on such samples. Artificial edge defects, 0.75 mm deep and 1.5 mm long, have been routinely detected by this system both when the positioning mechanism was used, and when the sample was hand-held in the inspection position. However, it is only recently, with the use of the positioning mechanism, that other defect areas have become apparent in the flat fuel sample. These appear to be caused by nonuniform distribution of UO_2 particles. These defects are almost impossible to detect with this system unless the sample is moving.

Further studies with the positioning mechanism are planned in order to determine better inspection speeds with this system.

b. Correlation of Heat Transfer and Bond Quality. The correlation between ultrasonic transmission capabilities of various types of mechanical bonds and their heat-transfer capabilities is being studied. To determine the thermal properties of the bonds, the thermal-pulse method for measuring diffusivity and conductivity developed by Parker et al.⁸ is being utilized. The front surface of the specimens is uniformly irradiated by a very short pulse (approx. 600 μsec) of radiant energy supplied by a xenon flash tube. The temperature history of the back surface is continuously monitored by feeding the output of a thermocouple in contact with the back surface into a differential amplifying system and then into an oscilloscope.

A satisfactory modification, enabling a constant and uniform contact to be maintained between the thermocouple and the specimen, has been made. A true back-surface temperature is recorded only when the resistance of the thermocouple and specimen circuit is at its lowest value. It was found that, for a 1- to 2-mm separation of the thermocouple leads at the specimen surface, this resistance has the same value for all materials and is equal to the resistance of the thermocouple itself. To

⁸ Parker, W. J., R. J. Jenkins, C. P. Butler, and G. L. Abbott, Flash Method of Determining Thermal Diffusivity, Heat Capacity and Thermal Conductivity, J. Appl. Phys. 32 1679 (1961).

insure the correctness of the temperature-rise measurements, this resistance value is checked each time a new contact is made.

Several measurements of diffusivity and conductivity of samples of copper bonded to copper with a silver-solder braze have been made. Each specimen was scanned over its central portion for about 30 thermocouple positions. Each position gives the diffusivity and conductivity in a 1-mm² area of specimen; readings are taken about 1 mm apart. Such closely spaced, point-by-point measurements are necessary, for poorly bonded specimens have been found to exhibit great differences of diffusivity and conductivity in small areas.

c. Application of Infrared Radiation to Nondestructive Testing. Investigation of the possible use of infrared in nondestructive testing has continued. A meter has been installed on the infrared imaging system for the purpose of providing a gray-scale calibration. When properly calibrated, the meter will indicate film density as a function of the beam intensity of the cathode-ray tube. This will aid in obtaining a properly exposed picture and facilitate adjustment of the black-to-white temperature difference.

The causes of poor temperature resolution are being investigated. The major problem seems to be the chopping motor and wheel. A 400-cps motor has been ordered which will allow a two-bladed chopper to be used instead of the present six-bladed chopper. This should eliminate the modulation due to slight differences in the blades of the chopper and still yield gain by a factor of two in chopping frequency.

Several pictures were taken with lead sulfide as the detector in the infrared imaging system. The signal-to-noise ratio was considerably better than that obtained at the same temperature with indium antimonide. Some smaller lead sulfide detectors have been ordered. They should further improve the signal-to-noise ratio as they will require no field stop.

d. Neutron Radiography. Nuclear fuels, when tested in reactors, soon become so radioactive that they must be encapsulated before the start of a test. The removal of specimens from their capsules (for examination) followed by the re-encapsulation of the specimen for further irradiation, is complicated, time-consuming, and usually detrimental to the success of the experiment. It is therefore advantageous to follow the experiment by photographing images of the specimens and associated equipment in the capsule. A method of obtaining the images by neutron radiography has been developed. The irradiated contents of three stainless steel fuel-capsules were recently observed in this way while still encapsulated. The specimens thus examined, CP-33, CP-34, and CP-35, are described in Section II.B.2 of this report.

e. Ultrasonic Propagation in Liquid Metals. Studies of ultrasonic propagation in liquid metal systems have been initiated in the Liquid Metals Laboratory in the Reactor Engineering Division. By these studies it is hoped to develop a method of indicating the depth of liquid metal in a container. A flat-bottomed stainless steel pipe has been inserted into a sodium tank and has been used as a receptacle for the ultrasonic transducer. With an oil couplant and a Saureisen cement-backed transducer, good ultrasonic echoes have been received from the bottom of the tank through several centimeters of sodium. A rough measurement of ultrasonic velocity made by moving the stainless steel pipe a known distance in the liquid sodium (at a temperature of about 175°C) yielded a value of 2.4×10^5 cm/sec, in reasonable agreement with published results.⁹⁻¹¹

Further effort to use transducers backed with other materials will be made. Improved backing designs are expected to reduce pulse lengths and permit more accurate measurements.

E. Engineering Mechanics

1. Grid Structure Experiment

The repetitive nature of reactor grid supports consisting of two circular plates interconnected by many tubes to carry fuel elements was subjected to a detailed analytical study, after which an experimental investigation was started to verify certain of the basic assumptions in the analysis.

A model to simulate the actual problem was relatively easy to fabricate. It consisted of two circular plates, 3 in. apart, connected by nineteen rods which were distributed in a hexagonal manner with respect to the center. A heavy armor plate was chosen to support the test model at the periphery of the plates. The same rigidity had to be provided for the support of both plates.

The grid model was so rigid that gravity loads were inadequate to produce measurable deflections. Application of nineteen individual loads above each rod was accomplished by the use of load rings made from standard pipe and calibrated on an accurate tensile tester. The load rings were mounted on a flat plate which in turn was supported by four posts from the base support (see Fig. 31). The picture also shows the top surface of the top plate of the model which is mounted inside the supports. Load rings, supported from above, bear against the upper plate with adjustable screws. The screws were used to apply the loads.

⁹Kleppa, O. J. Chem. Phys. 18, 1331 (1950).

¹⁰Pochapsky, T. E., Phys. Rev. 84, 553 (1951).

¹¹Trelin, Yu. S., I. N. Vasilev, and V. V. Roshchupkin, Soviet J. of Atomic Energy, 9, 955 (1960).

Figure 31
Comparison of Radial (σ_r) and Circumferential (σ_θ) Stresses in the Plates of the Grid Structure between Experimental Results and Analytical Predictions.

After modifications were made in the auxiliary supporting equipment to insure its rigidity, consistency in experimental readings was attained. Strain readings of the two plates and the rods, and deflections at the edge of the plates, were recorded for several sets of loads (250, 500, 750, and 1000 lb/each ring). Stresses in the plates obtained from experiments are compared with stresses predicted by analysis and shown in Figures 32-35. For comparison, compressive and tensile stresses are plotted on the same graphs.

Figure 32
Loading of
250 lb/ring

Figure 33. Loading of 500 lb/ring

Figure 34. Loading of 750 lb/ring

Figure 35. Loading of 1000 lb/ring

There are several important observations that are borne out in the graphs:

(1) At the edge, the magnitudes of stresses of the top and bottom plates are quite different. It appears that in the vicinity of the edge, the two plates do not deform identically. This difference, however, seems to be of such a magnitude as not to be noticeable in the readings of mechanical dial gages.

(2) The numerical difference between the top (compressive) and the bottom (tensile) stresses of each point of a plate is very small. This indicates that the neutral surface of each plate is at about half of the plate thickness. Thus, the assumption that each plate deforms independently is verified. The statement also implies that membrane stresses are negligible. This was another assumption made in the analysis.

(3) Strain gages closest to the center of the plates gave consistent readings continuously, yet these readings definitely show a deviation from the predicted values. There is no apparent reason at this time why the difference exists.

It must be borne in mind that the theoretical analysis of the grid structure is based on the assumption that the number of tubes or rods between the plates is large. The present model, however, deviates considerably from this assumption. It is quite important to observe such reasonable overall agreement between theory and experiment. Evaluation of the test results is continuing.

F. Heat Engineering

Following is a summary of that portion of the Laboratory's heat engineering program supported by the Division of Reactor Development.

1. Hydrodynamic Stability

The problem of flow stability in natural-circulation boiling systems was approached in two ways: (1) experimental measurements of power-to-void transfer functions, and (2) experimental measurement of the highest steady-state power that a given system can deliver. Both of these studies have been completed and reported: H. Christensen, Power-to-Void Transfer Functions, ANL-6385, (July 1961) and R. P. Anderson et al., An Analog Simulation of the Transient Behavior of Two-phase Natural Circulation Systems, ANL-6653 (Dec 1962).

Further work on hydrodynamic stability studies is being pursued as part of the ANL-AMU heat transfer program (see item 6e, Page 72).

2. Boiling Liquid Metal Studies

The primary objective of this program is to gather experimental information pertinent to the two-phase hydrodynamic and boiling heat transfer behavior of the alkali metals. The program is limited to the accurate measurement and correlation of fundamental parameters, and it does not encompass the development of vapor-cycle power systems, except as regards the use of components and materials necessary to the operation of experimental equipment.

The parameters of interest include two-phase pressure losses, variation with pressure and quality of the vapor volume fraction, boiling heat flux variations, condensing coefficients, and boiling stability. These parameters will be varied in a wide temperature range extending to 2100°F, and most experiments will utilize sodium as the fluid medium in forced-convection systems.

The successful completion of these tasks is highly dependent upon the materials selected for loop fabrication and the development of dependable high-temperature instrumentation. The materials experience of outside organizations who are developing power systems has been utilized.

The foregoing considerations have dictated that the experimental effort be divided into two overlapping phases.

The first phase consists of the design and operation of a forced-convection, boiling-sodium loop for the measurement of two-phase pressure losses (isothermal) and vapor volume fraction, stability studies, and instrumentation development. This facility is constructed of 316 stainless steel and is limited to short-term operation at temperatures between 1600 and 1700°F. Considerable data will be collected at temperatures below 1600°F (less than atmospheric pressure).

Construction of the loop is complete, and full operation awaits the installation of the inductive level probes. However, the following information has been obtained in preliminary studies prior to full loop operation.

A stability test of the two manometer level gages is being conducted in a bench test prior to installation in the loop. After two weeks of operation the output suddenly fell to zero. The difficulty was finally traced to a corroded ground connection in the recorder and has been corrected.

Calibration of the condenser level probe and the two probes required for differential measurement in the two-phase, isothermal pressure drop section is underway.

It was found through considerable experimentation that a simple "scrubber" principle (bubbling bottled argon through hot sodium directly into the loop gas control system) is not adequate for removal of water vapor and oxygen from the gas. The scrubber accumulates oxide rapidly and must be accessible for frequent cleaning; also, the system must be designed to prevent carryover of sodium vapor into cold, inaccessible portions of the gas control system. An external argon clean-up system has been installed. This system consists of two drying columns (containing magnesium perchlorate), a sodium bubbler, a cold tank for trapping sodium vapor, and a clean gas storage tank.

The measurement of pressure differentials (for flow rate and two-phase pressure loss) and the control of condenser liquid level is dependent upon the reliable operation of the 10-kc eddy-current liquid-level probe. This probe is composed entirely of alumina and chromel wire, and will operate in an air environment to 2100°F. The difficulties encountered in the read-out circuits have been overcome, and final calibrations are being carried out for all five probes.

The information obtained in, and the instruments developed for, the first phase of the boiling liquid metal studies will be applicable to the second general phase of the program. This second phase consists of the design and operation to 2100°F of a forced-convection, boiling-sodium loop,

with main emphasis on the measurement of the variation of the boiling heat flux with pressure, temperature, and the temperature difference between the heater wall and the saturation temperature of the fluid. This loop will be constructed mainly of Nb-1% Zr alloy and must operate in a vacuum environment at a total pressure of less than 10^{-6} torr.

The vacuum chamber for the loop is now under construction. Fabrication drawings of major components of the vacuum chamber have been reviewed and approved. Construction of the vacuum equipment is ahead of schedule. Rough assembly of both parts of the tank has been completed and the coolant lines are now being attached. The chamber supports and the water and liquid nitrogen baffles are completed. Work is well underway on the oven and control panel housing.

The water-cooled bus bar to the temporary load for the 150-kW power supply developed leaks and had to be replaced. Work on the check-out of this supply has been proceeding on a low priority basis in favor of initiating efforts on longer range aspects of the program.

The loop facility is under design. The main problem to be solved is the development of a high heat flux heater (up to 10^7 B/hr-ft²). It has been predicted that nucleate boiling superheats of several hundred degrees may occur during the initial operation of the heater, which indicates that the heater must withstand severe temperature conditions.

Small-scale tests of both radiant heaters and electron bombardment heaters are underway to solve fabrication difficulties and the problems encountered in measurements of power and temperature at the high temperatures involved. These tests are being conducted in a small, bench-scale stainless steel system designed simply to provide a flowing stream of hot liquid metal. An argon-purification system has been connected to the stainless steel loop in preparation for radiant heater tests. A power supply for driving the EM pump has also been installed. The problems involved in locating thermocouples in the radiantly heated test section have been resolved, and tantalum radiation shields have been assembled for use in the heated section.

Design of the small-scale electron bombardment heating experiment has been completed. The engineering drawings have been completed and approved for fabrication.

A critical review of this second phase of the boiling liquid metal study program has indicated that completion of the Nb-Zr loop will probably be delayed beyond the original target date, owing to technical difficulties and lack of manpower. The new target date for completion of this loop is the middle of 1965.

3. Heat Transfer in Double-pipe Heat Exchangers

A detailed analytical study of heat transfer in double-pipe heat exchangers, begun at the end of 1962, continues. In general, the method of attack consists of determining temperature and heat flux distributions from the basic laws of energy conservation and heat transport, rather than by use of heat transfer coefficients as in customary design calculations. In many situations, and with liquid metals and laminar flows in particular, the customary use of heat transfer coefficients is often of questionable validity. The main purposes of this study are (1) to investigate the dependence of actual heat transfer coefficients on the operating conditions of the exchanger, and (2) to examine the accuracy of current design calculation procedures as applied to liquid metal and laminar flow heat exchangers.

In the past year studies have been completed for various special cases as described in previous Progress Reports.¹² Currently, the effort in this study is being devoted to analyses of the cocurrent laminar-flow case, and to designing an experiment for verifying certain unusual effects predicted by the analysis for cocurrent turbulent liquid metal flows.

4. Fog-flow Heat Transfer and Fluid Flow

The heat transfer and fluid flow behavior of a highly dispersed mixture of liquid in its own vapor, or fog, has received considerable attention during the last few years, especially with respect to reactor applications. Of particular interest is the occurrence of a critical heat transfer condition in a fog flow, which appears to be similar in many respects to "boiling burnout" in boiling flows.

An attractive idealization of a real fog flow capable of reasonably exact mathematical representation was developed in 1962¹³ and is being developed further at ANL. The purpose of this research is to develop an analytical formulation with which the heat transfer and fluid-flow behavior of a fog flow can be predicted. Although originally intended for application to high-quality "boiling" water flows, the idealizations appear even more plausible when applied to the vaporization and condensation of liquid metals flowing through ducts.

¹²See, for example, Progress Report for January 1964, ANL-6840, p. 68; Progress Report for October 1963, ANL-6801, p. 53; Progress Report for April 1963, ANL-6717, p. 49.

¹³R. P. Stein et al., Investigations of West Steam as a Reactor Coolant (CAN-2), Final Report, Volume I, Behavior of a Fog Flow as a Coolant, UNC-5008-I, NYO-9844-1 (Aug 10, 1962).

Current efforts are being devoted to studying the results of applicable experiments being performed in England, as well as to extending previous mathematical formulations.

5. Boiling Heat Transfer Review

Research in various aspects of boiling heat transfer and fluid flow important to reactor technology is being reviewed on a continual basis. Emphasis has been placed on the indexing of "Boiling Burnout" data sources for use by reactor engineers. A first edition of such an index was completed for publication in the book Selected Topics in Reactor Technology, 1963 to be issued by the USAEC in 1964 (TID-8540). The index serves as a means of locating experimental data obtained with operating conditions and equipment closely duplicating a particular system being designed. Current efforts include the preparation of a second more complete and up-to-date edition of the "Boiling Burnout" data source index.

6. ANL-AMU Program

Other heat engineering experiments have been performed as part of a joint program (not supported by the Division of Reactor Development) between the Laboratory and the Associated Midwest Universities (AMU).

a. A Study of Two-phase Flow Characteristics during the Complete Vaporization of a Fluid. The complete vaporization of a fluid in forced-convection flow is being studied to obtain a map of the various flow patterns, a map of the void fraction, and the associated overall pressure drop for system pressures varying from moderate to the critical pressure. A range of mass flowrates will be employed as a parameter. A system has been designed to incorporate an electrically heated test section where visual observations can be made, and an unheated test section of identical shape will be installed at the exit of the heated section. A comparison of the void fraction and pressure drop between the heated and unheated sections is to be made.

The loop modifications are well under way. The existing pre-heater units have been disassembled, inspected and cleaned. Replacement and modification of some of the elements is being undertaken. Fabrication of the heat exchanger equipment and the modification of the existing test section has started. A burnout detector is being designed, and the design of equipment for the void-fraction measurement is nearly complete.

A correlation of the density ratio of saturated liquid to vapor for the Freons and water has been obtained on a reduced-pressure basis for the pressure range from 50% of critical to the critical pressure. This will enable a correlation of test results for the two systems. The surface tension of the Freons has been expressed as a function of the saturation.

A correlation of the viscosity and thermal conductivity is being attempted. The results of this analysis should provide a basis for correlation of test results on a dimensionless-group basis.

The test program should start within two months.

b. Transient Behavior - Heat Transfer Loop. The unstable behavior of a heat transfer loop when the coolant approaches its thermodynamic critical point is being studied. Previous work¹⁴ with a natural-circulation loop led to the hypothesis that the instabilities occur when the coolant bulk properties are in the thermodynamic region such that the product of the density and enthalpy is a maximum. This hypothesis is being tested by comparing the previous results with those obtained in a forced-convection loop. Investigation thus far indicates that the criterion of a density-enthalpy maximum is unsatisfactory.

A heat transfer loop has been assembled which can operate by either natural or forced convection. Several significant experimental results obtained therefrom are:

1. Sustained pressure oscillations can occur outside of the region that has been described previously. They have been encountered at bulk temperatures substantially lower than reported.

2. It is possible to move into or out of the unstable region while holding the bulk properties approximately constant by varying the flow rate, which in turn causes variation of the heater wall temperatures. From a given unstable position, stability may be achieved by either raising or lowering the flow rate, consequently decreasing or increasing the heater wall temperatures. The preliminary results indicate that boundary-layer mechanics are the controlling factor of the oscillations which have been observed. Possibly a boiling-like mechanism is involved.

Modifications are being made to permit better measurements of the wall temperatures and flow rates. It has not been possible to obtain satisfactory records of the wall-temperature history. The measurements to date have been made with nonrecording potentiometric-type instruments. A turbine flowmeter is being installed to permit better evaluation of the flow transients, since there is some question as to whether differential pressure oscillations measured across the Venturi are caused entirely by flow fluctuations.

c. Propagation of Void Waves in an Air-Water System. An electrical-resistivity probe will be used to investigate the propagation of void waves in an air-water natural-circulation system. To check reliability

¹⁴D. G. Harden, ANL-6710 (May 1963)

of void-fraction measurements by use of the probe, results of such measurements were compared with those from the X-ray attenuation technique. The probe measurements resulted in low values of void fraction¹⁵

The probe was made sharper. High-speed movies were taken of the piercing of an air bubble in water by the probe. The resulting electrical signal was superimposed on the same film in order to note the response. It appears that the response from the probe is instantaneous. Also, there is no apparent deflection of the air bubbles by the probe.

On the basis of these movies, void-fraction measurements from the improved probe were checked by comparison with results from (1) X-ray attenuation, and (2) pressure drop. These tests took much longer to run than was predicted. The cause was traced to faulty functioning of the electronics for the density equipment. The equipment was modified and is now working properly. Additional data are being taken.

d. Power-to-Void Transfer Function Measurement. In order to study the power-to-void transfer functions in a channel, the power to a single, out-of-pile, coolant channel will be modulated sinusoidally, and the transient response of steam in the channel will be analyzed by a cross-correlation technique. The operating conditions that have been chosen are:

a) Low inlet subcooling to give a well-defined bulk-boiling region where there is complete transverse mixing.

b) A thin-walled test section to decrease heat exchange mechanisms between coolant and wall.

Modifications of the 2000-psi loop are essentially complete, and several preliminary runs have been made. A new test section with a $\frac{1}{64}$ -in. wall thickness has been obtained, as well as new end pieces and new backup plates. In order to facilitate finer temperature control of the inlet stream to the test section and to minimize the inlet subcooling, the existing line heaters on the loop have been repositioned and a new bayonet-type Calrod heater installed. In order to reduce pressure feedback from the steam separator, a larger tank has been installed.

e. Heat Transfer to Water under Supercritical Pressures and Natural-circulation Conditions. The supercritical loop has been completed and tested at pressures to 4000 psi. Several preliminary tests have been performed at pressures to 3400 psi with powers up to 35 kW. A gasket failed and is now being replaced. A number of "bugs" in the loop operation and response were found and are now being corrected:

¹⁵G. P. Nassos, ANL-6738 (June 1963).

a) The loop pressure response was unsatisfactory and is being modified by introducing an accumulator in the system.

b) The pressure drop across the Venturi was found to be too high in comparison with the total pressure drop in the loop, and a new Venturi meter with a larger throat section has been designed and ordered.

c) Extensive heat balances indicate that a large amount of power is lost through the copper electrode bars. Additional thermocouples are being installed to establish the heat losses more accurately.

d) The temperature at the top of the riser was found to be higher than at the outlet of the test section. This is thought to be due to a very sharp change of temperature distribution across the pipe at the outlet of the heater. The thermocouple at the top of the test section has been changed in order to check this fact.

f. Inception of Hydrodynamic Instability in a Natural-circulation System. Instability data accumulated from previous studies have been analyzed. The 28 data points used had been obtained at five different pressures, for three test-section diameters, and for eleven different combinations of restrictions in riser and downcomer. Because of the diversity of these data, no conclusions could be drawn about the effect of any parameter on the inception of instability. Further experiments seem necessary.

To obtain the additional data on the inception of instability in a natural-circulation loop, the Armadilla loop was modified for 1500-psi operation. A 50-kva cooler was added to the loop for a precise control of inlet subcooling. The modifications of the loop are nearly complete. The following is the range of parameters intended to be covered for this study:

1. Pressure	200 to 1500 psi
2. Inlet Subcooling	0 to 50°F
3. Test-section Diameter	$\frac{3}{8}$, $\frac{5}{8}$, $\frac{7}{8}$ in.
4. Test-section Length	72, 96, 120 in.
5. Riser Diameter	$\frac{5}{16}$, $\frac{5}{8}$, $1\frac{1}{4}$ in.
6. Riser Length	40, 50, 60 in.

In addition to the experimental program, concurrent theoretical analyses are being carried out.

G. Chemical Separations

1. Chemistry of Liquid Metals

Thermodynamic data have been calculated for the intermediate phases of the system lanthanum-cadmium by means of equilibrium pressure

measurements made over the two-phase regions LaCd_{11} - $\text{La}_2\text{Cd}_{17}$, $\text{La}_2\text{Cd}_{17}$ - $\text{La}_3\text{Cd}_{13}$, $\text{La}_3\text{Cd}_{13}$ - LaCd_2 , and LaCd_2 - LaCd (see Progress Report for January 1964, ANL-6840, p. 71). The value for the free energy of formation of LaCd_{11} at 500°C had previously been determined to be -3.22 kcal/g-atom in galvanic cell studies (see Progress Report for December 1963, ANL-6810, p. 36). The standard free energies of formation at 500°C of the remaining intermediate phases: $\text{La}_2\text{Cd}_{17}$, $\text{La}_3\text{Cd}_{13}$, LaCd_2 , and LaCd , were calculated from the free energy value for LaCd_{11} and from the vapor pressure data. Tentative values are: -3.91, -6.30, -7.89, and -7.42 kcal/g-atom, respectively.

2. Fluidization and Volatility Separation Processes

a. Recovery of Uranium from Highly Enriched Uranium-Alloy Fuels by Chlorination and Fluorination Steps

(i) Laboratory-scale Studies. Studies of the feasibility of a 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-aluminum and uranium-Zircaloy fuels in a $1\frac{1}{2}$ -in.-diameter fluid-bed reactor with the alloy subassembly submerged in an inert bed. An additional run using multiplate uranium-Zircaloy subassemblies was carried out (see Progress Report for October 1963, ANL-6801, p. 61) (1) to test the behavior and uranium retention of sintered alumina in the fluid-bed reactor and the packed-bed filter, (2) to determine the disposition and distribution of the minor constituents of Zircaloy-2 and selected simulated fission products added to the fluid bed, and (3) to determine the effectiveness of a sintered Monel filter interposed between the packed-bed filter and the cold traps for retaining particulate material during fluorination.

In this run, the charge consisted of two 137-g uranium-Zircaloy-2 subassemblies (1.9 w/o normal uranium), each subassembly consisting of three plates and being 5 in. long. In addition to the fuel subassemblies, 13.7 g 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 sintered alumina (Tabular type T-61; Aluminum Company of America). The fluid bed was composed of -30 + 60 mesh alumina particles; the filter bed was composed of -14 + 20 mesh alumina particles.

The following reaction sequence was employed in the run: hydrochlorination, hydrofluorination, and fluorination. The hydrochlorination step, with moist hydrogen chloride (1100 ppm water), was conducted for 8 hr at fluid-bed temperatures which ranged from 339 to 515°C and at a

packed bed temperature of 350°C. The hydrofluorination step with 30 v/o HF in nitrogen was conducted at a fluid-bed and packed-bed temperature of 350°C over 2 hr. The fluorination step was carried out in two periods of 2 hr at 250°C and 2 hr at 500°C. In each fluorination period, the concentration of fluorine was gradually increased from 5 to 90 v/o (remainder nitrogen) during the first $1\frac{1}{2}$ hr and remained at the higher concentration during the last $\frac{1}{2}$ hr. The sintered Monel filter (of 10- μ pore size) was maintained at a temperature of 200°C during the fluorination periods.

The sintered alumina was found to be a satisfactory inert bed material. The alumina in the fluid bed remained fluidizable throughout the reaction cycle. The alumina in the packed-bed filter was freely flowing at the end of the reaction cycle. The retentions of uranium by the alumina in the fluid-bed reactor and the packed-bed filter were very low, 0.008 w/o and 0.006 w/o, respectively.

The relative distributions of most of the minor constituents of Zircaloy-2 and of the fission products conformed to the distributions expected from the volatilities of their higher valent chlorides and fluorides. These data are in agreement with the results obtained in a previous run (see ANL-6801, p. 61).

The four major elements found with the UF_6 in the cold traps were (expressed as percent of original amount charged): molybdenum, 93%; zirconium, <0.1%; ruthenium, 10%; niobium, 17%. The presence of zirconium in the product cold traps suggests that not all of the zirconium fluoride in the UF_6 process gas stream may be filterable.

Although the Monel metal filter did not effectively retain zirconium for which it was primarily installed, it was effective for the removal of ruthenium. Additional runs are planned with the filter; the temperature of the filter will be lowered, probably to 60°C, in an effort to improve the retention of particulate material by the filter.

b. Pilot-plant Demonstration Runs. An additional shakedown run has been completed in the pilot-plant facility installed to demonstrate the recovery of uranium from highly enriched uranium-alloy fuels by means of the fluid-bed fluoride volatility process (see Progress Report for December 1963, ANL-6810, p. 38). In this run, a multiplate, welded sub-assembly which was fabricated from aluminum only was charged to the fluid-bed reactor. Since the subassembly did not contain uranium, only the hydrochlorination and hydrofluorination reaction steps were carried out. (In future experiments with subassemblies containing uranium, the fluorination step will be included in the reaction sequence.) During the hydrochlorination step, the fluid-bed pyrohydrolysis reactor was in operation to convert the volatile aluminum trichloride, which represents a waste stream, to a solid oxide.

The objective of this run was to investigate the effect of high concentrations of hydrogen chloride (in nitrogen) on the hydrochlorination reaction rate for aluminum and the utilization efficiency for hydrogen chloride.

The charge consisted of a 14-plate aluminum subassembly which was $2\frac{1}{2}$ in. by $3\frac{1}{4}$ in. by 48 in. long and weighed 12.3 kg. The reaction bed contained 39.6 kg (static bed depth of 46 in.) of -28 +100 mesh alumina;* the packed-bed filter contained 9.0 kg (bed depth of $6\frac{3}{4}$ in.) of -14 +28 mesh alumina.* The starting bed material for the fluid-bed pyrohydrolyser was 20.0 kg (static bed depth of 22 in.) of aluminum oxide (+100 mesh) remaining from the pyrohydrolysis of aluminum trichloride vapor from a previous run.

The hydrochlorination step was performed over a period of 6.4 hr at an average reaction bed temperature of 290°C. In this step, the hydrogen chloride concentration was 80 v/o HCl in nitrogen for the first 35 min of the feeding period; it was then increased to about 99 v/o HCl in nitrogen for the remainder of the hydrochlorination period. The hydrofluorination step (20 v/o HF in nitrogen) was conducted at a fluid-bed temperature of 260°C for 0.8 hr. The packed-bed filter was operated at 260 to 300°C and the fluid-bed pyrohydrolyser was operated at 300°C.

The results of previous runs (see Progress Reports for November and for December 1963; ANL-6808, p. 57, and ANL-6810, p. 38) have indicated that the hydrochlorination reaction rate increases with increasing hydrogen chloride feed rate and that the overall utilization efficiency for HCl decreases with increasing HCl feed rate. In the current run, during the first 2.5 hr of operation, a reaction rate of 2.6 kg aluminum/hr was achieved. This rate was higher than the reaction rates of 2.0 and 1.4 kg aluminum/hr achieved during similar periods in previous runs (see ANL-6808, p. 57, and ANL-6810, p. 38). The high rate achieved in the current run is due to the higher concentration of HCl in the feed, 99 v/o HCl in nitrogen as compared with 80 and 55 v/o HCl in nitrogen used in the two previous runs. The overall reaction rates in the current run and the previous runs were 1.9, 1.5, and 1.2 kg aluminum/hr, respectively. In the two previous runs, 14- and 17-plate aluminum-only subassemblies ($2\frac{1}{2}$ in. wide by 48 in. long) were charged to the reactor.

In the current run, the overall HCl utilization efficiency was 45%, which may be compared with overall utilization efficiencies of 51% and 55% for the two previous runs. The utilization efficiency appears to decrease with time, which implies that the utilization of HCl may be dependent on the length of the subassembly or on the amount of subassembly that remains unreacted. Utilization efficiencies as high as 80 to 90% were achieved for short periods during the first portion of the hydrochlorination

*Tabular Type T-61 sintered grain alumina, a product of Aluminum Company of America.

period. The overall utilization efficiency of HCl would probably be improved by the addition of a new subassembly toward the end of a batch hydrochlorination period. This effect will be investigated in a future experiment. A maximum temperature of 410°C was observed in the channels between the aluminum plates of the subassembly during the hydrochlorination reaction step; this temperature is well below the melting point of aluminum (660°C).

The aluminum chloride produced in the hydrochlorination reaction step was converted to the oxide in the fluid-bed pyrohydrolysis reactor. A major fraction (about 80 to 90%) of the aluminum oxide was found to be in the form of fines (3 to $30\ \mu$). This quantity of fines affected the operation of the pyrohydrolysis reactor in that the aluminum oxide automatic take-off valve plugged. An improved continuous overflow system has been installed and satisfactorily tested.

3. General Chemistry and Chemical Engineering

a. Reduction of Uranium Ore Concentrate. The direct reduction of uranium ore concentrate to metallic uranium, first studied with Anaconda resin-in-pulp ion exchange concentrate, has been extended to include nine additional concentrates that are typical of current feed materials: Union Carbide Nuclear ammonium diuranate concentrate, Union Carbide Nuclear sodium and/or ammonium diuranate concentrate, Vanadium Corporation of America concentrate of unknown type, Western Nuclear ammonium diuranate concentrate, Atlas Corporation magnesium ammonium diuranate concentrate, Texas Zinc Minerals magnesium diuranate concentrate, Phillips Petroleum sodium diuranate concentrate, Homestake Sapin sodium diuranate concentrate, and Kermac Nuclear concentrate of unknown type. The procedure was as follows:

1. removal of 80 to 90 percent of the silicon by contacting the ore concentrate with ammonium bifluoride (NH_4HF_2) for 4 hr at 150°C and removing the excess NH_4HF_2 by heating to 600°C . (Silicon removal may be considered an optional step, since silicon can be removed by the producer of the concentrate. Therefore, this step was performed only for three concentrates.);

2. chlorination of the concentrate by adding MgCl_2 and heating to 800°C in air for 2 hr with agitation. Oxidation of MgCl_2 by the air releases chlorine, which is then available to form volatile chlorides of impurity elements;

3. reduction of the concentrate to metal by zinc-magnesium alloy, using a MgCl_2 flux;

4. retorting of the resulting zinc-magnesium-uranium alloy under vacuum to remove the zinc and magnesium;

5. consolidation of the uranium sponge from the retorting step by arc melting.

Results of demonstration runs (on a 15-g uranium scale) with the ten concentrates indicate the following:

The reduction yields ranged from 95 to 99%. Overall uranium recoveries in these small-scale runs, however, ranged from 70 to 96%; the lower values resulted mainly from incomplete pouring of the liquid metal solution. It should be noted, however, that the heel remaining in a reduction crucible after pouring is recovered, since the crucible is reusable.

Very efficient removal of the alkali and alkaline earth metal impurities was achieved in this process. These impurities were present in concentrations up to 8 w/o in the ore concentrates, whereas in the uranium product the concentrations of barium, calcium, strontium, and potassium were below the limits of detection (below 10 to 50 ppm) and the sodium concentration in the product was 10 ppm or less.

In the uranium metal products from the ten concentrates, the concentrations of chromium, manganese, nickel, silver, and copper generally met the requirements for reactor-grade metal. The concentrations of silicon, iron, and phosphorus in the product exceeded specifications by one order of magnitude, that of vanadium and molybdenum by two orders of magnitude, and that of boron by two to three orders of magnitude. Zirconium, boron, and tantalum concentrations in the product were higher than in the concentrate as a result of contamination from the equipment and from impurities in the reagents. In the vacuum retorting and arc-melting steps, magnesium concentration was lowered to 10-35 ppm and zinc concentration was lowered below the limits of detection.

Further development work would be required before this process could be used to produce reactor-grade uranium metal. However, no further work on this procedure is planned at this time.

b. Preparation of Uranium Monocarbide. Refractory compounds of the actinide elements show promise as reactor fuels capable of withstanding high temperatures and large burnups. Particulate uranium monocarbide is being prepared on a 0.5-1 kg scale by the Chemical Engineering Division for evaluation as a fuel material by the Metallurgy Division. Three additional batches of uranium carbide have been prepared by adding carbon to uranium dissolved in zinc-12 to 14 w/o magnesium alloy at 800°C and stirring at 350 to 400 rpm for 8 hr. The zinc-magnesium solution is removed by pressure-siphoning followed by sublimation at 600-850°C under reduced

pressure. Different sources of carbon were used in the three runs - graphite lubricating powder (49% of the powder had a particle size of less than $15\ \mu$), charcoal black (average particle size of $70\ m\mu$), and activated charcoal (average particle size less than $44\ \mu$). The characteristics of the uranium monocarbide product (see Table XV) may be compared with the desired characteristics which are: (1) carbon content near the theoretical content of 4.80 w/o (2) oxygen content below 0.20 w/o, and (3) particle size of 1 to $10\ \mu$. Although a satisfactorily low oxygen content has been made, additional studies are needed to achieve all three requirements. These studies will be made with graphite powder and activated charcoal as carbon sources.

Table XV. Uranium Monocarbide Production Runs
(500- to 1000-g UC scale)

<u>Conditions</u>				
Reaction Temperature: 800°C				
Mixing Speed: 350-400 rpm				
Solvent: 12-14 w/o Mg in Zn				
<u>Carbon Source</u>	<u>UC Product</u>			
	<u>Total C (w/o)</u>	<u>Oxygen (w/o)</u>	<u><44 μ (w/o)</u>	<u><149 μ (w/o)</u>
Graphite Powder ^a	7.80	0.37	~67	-
Charcoal Black ^{a,b}	3.99	0.16	-	28
Activated Charcoal ^c	5.59	0.14	-	-

^a24-hr run.

^bCharcoal black proved unsatisfactory. Because of its fineness, it was easily removed from the crucible by gas currents, leaving the product carbon-deficient.

^c8-hr run.

c. Evaluation of Molten Nonmetallic Fuels for Fast Plutonium Breeder Reactors. Two of the potential advantages of mobile fuels for fast plutonium-breeder reactors are their high coefficient of thermal expansion, which may contribute to reactor safety, and the elimination of the decladding and fuel fabrication steps required by solid fuels. On the basis of literature data and calculations pertaining to reactor size, heat transfer problems, and required fuel solubility, it was concluded that the

most promising molten nonmetallic fuels consist of chlorides of plutonium and uranium diluted with other chlorides to lower the melting point. With this type of fuel, it is thought that internally cooled reactors having 10,000- to 20,000-liter cores could produce 1000 MW(e) of power with an estimated breeding ratio of 1.2 to 1.5 and a doubling time of about 15 yr.

The chlorine balance in a $\text{PuCl}_3\text{-UCl}_3$ fuel as a function of burnup was assessed from an estimate¹⁶ of the fission product concentrations for fast-neutron irradiation of Pu^{239} and from an evaluation of the most stable valence states of these fission product elements in the expected reactor environment. The average valence of the fission product elements was compared with the valence of the plutonium atom prior to fission. Indications were that there would be too few chlorine ions to satisfy the requirement of electroneutrality and that noble metals such as ruthenium and palladium would be present as metals in the expected reactor environment. Study of thermodynamic data indicated that some of the fission product chlorides could electroplate on the container walls in a corrosion-type reaction, dependent upon the container materials used.

d. Mixing in Liquid Metals and Salts. Four additional runs (see Progress Report for October 1963, ANL-6801, p. 64) have been made in an investigation to correlate mixing power input with the rate of reduction of uranium oxide. In each of the recent runs, 194 g of skull oxide was suspended in $\text{MgCl}_2\text{-CaCl}_2\text{-MgF}_2$ flux and was contacted with zinc-magnesium alloy at about 800°C. As expected, the rate of uranium oxide reduction increased at greater mixing power inputs. In two runs at a mixing power input of approximately 10^{-3} hp, a potentially significant phenomenon was observed: 30 min elapsed before any appreciable reduction occurred, and then the reduction proceeded smoothly and rapidly. The reasons for this phenomenon are being investigated in order to determine how to avoid the delay period.

e. Distribution of Californium between Magnesium Chloride and Zinc-Magnesium Alloy. An experiment has been performed to check preliminary results (see Progress Report for November 1963, ANL-6808, p. 53) on the distribution behavior of tracer californium between magnesium chloride and magnesium-zinc alloy at 800°C. The distribution coefficients have been redetermined at 710°C. The results, which confirm the earlier data obtained by another technique, show that californium has an increasing tendency to distribute to the metal phase as magnesium concentration of the alloy is increased from 14 w/o to 66 w/o. This behavior differs from behavior in this solvent system of other elements investigated thus far (see Progress Report for April 1962, ANL-6565, p. 27).

¹⁶L. Burris, Jr., and I. G. Dillon, ANL-5742 (July 1957).

f. Materials and Equipment Evaluations. Crucible materials are needed which will satisfactorily contain molten zinc alloy and halide salt systems. Two 2-in.-dia tungsten pipes, plasma-coated on the inside with beryllia, have been subjected to preliminary thermal-cycling tests. One pipe withstood six thermal cycles between 250°C and 900-950°C in argon atmosphere. The second pipe withstood the action of 50 w/o zinc-magnesium solution held at 800°C for one hour, cooled, reheated to 800°C, and poured.

g. Head-end Treatments for Refractory Fuels. In the reprocessing of spent uranium monocarbide fuel by pyrometallurgical means, conversion of the fuel into a form that is soluble in molten chloride salts is desirable for subsequent processing steps. Previously (see Progress Report for November 1963, ANL-6808, p. 59), chlorination of the carbide with HCl in a molten chloride flux was shown to convert the carbide to soluble uranium chlorides and free carbon. For further reprocessing of the fuel, the carbon should be removed. Preliminary results indicate that bubbling SO₂ through a solution of UCl₄ in LiCl-KCl eutectic may remove the carbon without oxidizing the uranium chlorides to UO₂Cl₂. It is desirable that the uranium compound be nonoxygenated so that solid magnesium oxide does not appear in the salt in a later process step.

h. Decladding Studies. Scouting studies indicate that removal of TV 20 (a promising vanadium-titanium cladding alloy) from a fuel may be possible by chlorination with chlorine diluted with helium at 250°C. The tetrachlorides of both vanadium and titanium can be removed by distillation.

H. Plutonium Recycle Program

1. Errata in Report on Density Coefficients of Reactivity

In the Reactor Development Program Progress Report for January 1964, ANL-6840, p. 77, all of the water density coefficients of reactivity for the EBWR plutonium recycle experiment were incorrectly reported as negative. All of the density coefficients of reactivity are actually positive. If one replaces the word "density" with "void," then the negative signs would be correct as presented.

2. Plutonium Recycle Fuel

A three-zone fuel loading is being fabricated for the Plutonium Recycle Experiment. This loading is a joint effort of G. E. Hanford Atomic Products and Argonne National Laboratory. The three zones are as follows:

(a) central test zone of mixed plutonium oxide-uranium oxide. This zone contains 36 elements fueled with 1.5 w/o plutonium oxide and 98.5 w/o depleted uranium oxide;

(b) intermediate shim zone of enriched uranium with a rare earth burnable poison. This zone contains 60 elements fueled with UO_2 enriched to 6 w/o U^{235} . The burnable poison is 0.158 w/o Eu_2O_3 and 0.0288 w/o Sm_2O_3 ;

(c) blanket zone of natural uranium oxide. This zone contains 58 elements fueled with natural UO_2 .

All fuel elements contain 36 fuel rods in a square lattice. The fuel is contained in 0.945-cm-ID x 0.064-cm-wall Zircaloy-2 tubing.

Fuel rods for the central test zone are being loaded at Hanford by vibratory loading the mixed oxide into the Zircaloy-2 tubes. The mixed oxide is prepared by a high-temperature high-energy-rate forming process which produces a uniform dispersion of PuO_2 in UO_2 . This uniform dispersion is maintained throughout the vibratory compaction of the fuel.

In addition to the fuel rods required for the 36 central test elements, 216 rods are required for four spare elements and approximately 460 rods are required for physics tests at Hanford. All mixed-oxide fuel rods will be assembled into fuel elements at ANL.

All facilities for processing the fuel at Hanford are completed. Approximately 1,000 kg of the mixed oxide has been processed through the high-energy-rate forming operation. With the exception of several hundred feet of Zircaloy-2 tubing ordered as a replacement for rejected material, all hardware for the mixed-oxide fuel rods has been received. A total of 460 loaded rods have been tested and delivered for Approach to Critical Experiments and for loading in the Plutonium Recycle Critical Facility at Hanford. An additional 100 rods acceptable for Plutonium Recycle elements have been loaded.

The intermediate and blanket zone uranium oxide fuel rods and assembled elements are being fabricated by United Nuclear Corporation on Contract No. 31-109-38-1622. This material is being furnished in accordance with drawings and specifications prepared by the Laboratory.

United Nuclear has converted all the natural UO_2 to UO_2 pellets which have been inspected, weighed, and loaded into column lengths in preparation for loading into the blanket rods. All enriched UF_6 has also been converted to UO_2 and master blends of UO_2 with the required additions of rare earth oxide have been prepared. These blends are being analyzed for rare earth content and homogeneity before proceeding with the enriched pellet production.

All Zircaloy-2 strip stock, Zircaloy-2 tubing, Zircaloy-2 end plug stock, end fittings, grid castings, upper spacer grid, and miscellaneous hardware have been subcontracted by United Nuclear Corporation. Present schedules call for delivery of evaluation tubing in two weeks, end fittings in

one week, lower grid casting by March 2, upper grid casting by March 23, top spacer grid by March 10, and compression springs by March 10. All rod stock for end plugs is due in five weeks and all strip stock for fuel can fabrication is due in three weeks.

Present production schedules call for delivery of all fuel and hardware before the end of the second quarter of FY 1965.

3. Plutonium Recycle Control Rods

A total of eight new control rods containing 2 w/o boron enriched to 90% B¹⁰ in stainless steel will be fabricated in place of natural boron in stainless steel for the absorber section. One central hafnium oscillator rod is also being replaced.

The boron stainless steel is being fabricated by Carpenter Steel Company in accordance with specifications prepared by the Laboratory.

The replacement control rods have been redesigned to eliminate the mechanical riveted transition section between the absorber section and the 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.

Boron stainless steel and hafnium will be furnished by the Laboratory to commercial suppliers for the fabrication of the control rods.

4. Heat Transfer and Hydrodynamics

The final calculations concerning the thermal-hydraulic performance of the core have been made and will be incorporated into the Safety Analysis Report. Table XVI outlines the more important predicted characteristics at the initial power level of 42 MW. Since the core diameter and the riser and downcomer geometry will not change from previous EBWR operation, and because the equivalent hydraulic diameter is only slightly smaller than the average of previous cores, some of the experimental results of Core IA operation have been used to yield more accurate predictions of performance.

Table XVI. Predicted Performance at 42 MW

Reactor Power	42 MW
Operating Pressure	600 psig
Operating Temperature	489°F
Average Steam Void Fraction (based upon coolant vol)	0.262
Average Steam Void Fraction (based upon total moderator vol)	0.210
Core Power Density, avg. (based upon coolant vol) max	42.17 kW/ℓ 130.5 kW/ℓ
Total Recirculation Flowrate	7.51 x 10 ⁶ lb/hr

It should be mentioned that, since power production is not of primary interest for the recycle program, several sets of computations were carried out for power levels between 20 and 60 MW before a satisfactory compromise among power, void fraction, fuel composition, and isotopic distribution was evolved. Power-void iterations were carried out by two codes programmed for the IBM-704 computer: the PDQ physics code, and the CHOPPED (RE-293) hydrodynamic program. The CHOPPED code is a revision of RECHOP, previously used extensively for EBWR calculations. The new code provides a faster iteration procedure, and it was possible to produce relationships between local average void fraction and power level at a given burnup. It is calculated that the initial power level of 42 MW will decay with time to 30 MW at the end of irradiation.

There has been a proposal by Hanford that some special rod irradiations be performed in the plutonium zone of the recycle core; these rods would be composed of plutonium-aluminum alloy clad with Zircaloy-2. A few rods of this type have been irradiated in the PRTR at Hanford, and the thermal expansion difficulties are apparently solved by allowing a sufficient gap between fuel and clad at fabrication. Further aspects of the problem are being investigated.

5. Storage Rack Modifications

The reactivity resulting from storing plutonium recycle fuel in the EBWR south storage rack was calculated. When the south rack is loaded with fuel from the plutonium zone (which loses reactivity with irradiation), the assembly had a multiplication constant of 0.916 at zero burnup; when the rack is loaded with fuel from the shim zone (which gains reactivity monotonically with irradiation), the assembly had a multiplication constant of 0.964 with 0.006 a/o burnup. Since it is necessary absolutely to preclude the possibility of the loaded rack reaching criticality and since the calculated multiplication constant of the rack when loaded with fuel from the shim zone with 0.006 a/o burnup is near unity, it is desirable to modify the rack so as to increase its absorption.

The south storage rack presently consists of a rectangular array of 143 4-in. sq boxes. The rack was made by interlocking $\frac{1}{8}$ -in.-thick sheets of virgin stainless steel with $\frac{1}{8}$ -in.-thick sheets of 1 w/o boron-stainless steel to form boxes which have two borated and two virgin stainless steel sides. An economical way of increasing the boron content is to use 1.2 w/o boron-stainless steel for all sheets.

By reconstructing the rack and using 1.2 w/o boron-stainless steel for all sides, the multiplication constant for the most reactive case is estimated to be no more than 0.878 with $\frac{1}{8}$ -in.-thick sides and 0.811 with $\frac{1}{4}$ -in.-thick sides.

6. Shipping Cask Heat Transfer Analysis

The analytical study of transient cask temperatures (see Progress Report for January 1964, ANL-6840, p. 71) has been completed. The results indicate that the surface temperature limitation of 180°F will not be exceeded with a full load of seven EBWR "spike" assemblies and "maximum" solar heating. The allowable heat generation from the fuel under this solar heat load, however, is only a fraction of that anticipated from a single plutonium-bearing assembly. To avoid such a stringent limitation on future transport capacity it is recommended that 1) further consideration be given to providing fins on the cask and 2) the expected solar heating conditions associated with the individual shipment be examined in future calculations.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Core Physics

A careful recompilation of the 18-group cross section for three temperatures: 21°C, 85°C, and 100°C, has been concluded. The pure temperature coefficient of reactivity, $\partial \ln k / \partial T$, can be represented over the range from 21°C to 100°C by the function $(a + bT)$, where T is the temperature (°C) of the reactor (assumed uniform over the entire reactor). This coefficient is not to be confused with the total temperature coefficient

$$\frac{d \ln k}{dT} = \frac{\partial \ln k}{\partial T} + \frac{\partial \ln k}{\partial \ln \rho_W} C(T) + \frac{\partial \ln k}{\partial \ln V_M} 2\alpha_M + \frac{\partial \ln k}{\partial T}_{\text{Doppler}},$$

where ρ_W is the water density at temperature T , $C(T)$ is the water compressibility at T , $\partial \ln k / \partial \ln V_M$ is the percent reactivity change per percent metal (fuel) volume change, α_M is the linear coefficient of expansion of the fuel at T , and the last term gives the Doppler effect. All the last-named terms are negative.

The results show a very desirable power coefficient which tends to become more strongly negative the higher the power. The total temperature coefficient is also rate-dependent, for if the rate of change of power is rapid enough, the temperature response will vary across the reactor. For example, the reflector, which contributes positive reactivity feedback with temperature, will lag farther and farther behind the core in temperature the faster core temperature rises. This means the inherent shutdown coefficient becomes more strongly negative the faster the rise in power. For very fast excursions (where there is insufficient time for water heating), only the metal-expansion and Doppler terms survive. The general tendency of the power coefficient as a function of rate of power increase is shown in Figure 36.

Figure 36. General Tendency of Power Coefficient of Reactivity vs. Rate of Power Increase

To assist the detailed study of reactor dynamics with the analog computer, various spatially distributed perturbations have been calculated by the 16-group cross section set. Typical among these is the void coefficient, $\Delta k/k$ per cm^3 of void. An approximate graph of the results is given in Figure 37.

Figure 37. Distributed Water Void Coefficient

2. Critical Experiment

Following AEC approval of the fuel contract, a minor delay resulted because of uncertainty over the amount of UF_6 specified in the contract, since it involved conversion of UF_6 to uranium metal, as well as rolling of metal to dimension.

Study of means of irradiating foil materials and measuring reactivity effects of sample coupons in the critical experiment have resulted in proposals of specific locations which are now being evaluated in relation to physics requirements. The accessible locations are quite limited, and careful planning is required to obtain a reasonable selection.

A measurement of the amount of air entrapped in a dummy fuel element when immersed in water showed that about 3-4 v/o (or 30 cc) of air remained. The measurement will be repeated when fuel is available, since different results may be obtained with the actual components. Meanwhile, efforts will be made to reduce the entrapment by adding wetting agents to the water and/or de-aerating the water.

3. Heat Transfer

a. Core Analysis. A heat transfer analysis of the AARR core operating at 240 MW was made by the STDY-3 code to determine the power increase factor (PIF), which is a measure of the power increase necessary to cause flow instability resulting in autocatalytic burnout. The PIF was evaluated as a function of the uncertainties in the design variables. The nominal core design conditions assumed in the analysis are listed in Table XVII.

Table XVII. AARR Nominal Core Design Conditions

Power, MW	240
Core volume, liters	77
Heat transfer area, ft ²	718
Average velocity, ft/sec	40
Heated plate length, in.	18
Channel spacing, in.	0.040
Plate thickness, in.	0.040
Clad thickness, in.	0.005
Maximum/average power density, radially	1.95
Maximum/average heat flux, axially	1.28
Axial power distribution	chopped cosine
Inlet core pressure, psia	750
Hot channel exit pressure, psia	670
Inlet temperature, °F	135

Table XVIII presents the uncertainties used in the analysis. A "hot streak" analysis was used in which it was assumed that all the factors occur along the entire length of the fuel plate and no fluid mixing occurs. In the

Table XVIII. Uncertainties and Tolerances

		Reference ¹⁷ <u>Designation</u>
Channel spacing, in.	0.038-0.042	V ₁
Local channel spacing, in.	0.036-0.044	V ₂
Entrance and exit area ratio	0.475-0.525	V ₃
Average plate factor	0.785-1.274	V ₄
Local plate factor	0.738-1.355	V ₅
Flow rate factor	0.872-1.147	V ₆
Power density factor	0.925-1.080	V ₇

¹⁷ A. C. Nelson, Jr., and W. S. Minkler, A General Method for Evaluating the Effects of Uncertainties in Design Variables on Core Performance, Nucl. Sci. Eng., 17, 101-110 (1963).

analysis, each variable was changed in a prescribed manner to conform to an orthogonal composite design in which only one-fourth of the points on a hypercube are used.¹⁷ Consequently, 47 problems with different sets of values for each of the factors in Table XVIII were solved to determine the PIF. The PIF was expressed by first- and second-degree terms in each of the variables and their products in a polynomial determined by a least-squares fit. The results of the least squares empirical fit have been obtained, but have not been verified. An excellent fit ($\pm 0.2\%$) appears to have been obtained.

With these ranges of variables, PIF values between 1.25 and 2.23 were obtained, with a PIF of 1.68 for an entirely nominal core.

The ranges of variables must be extended beyond those shown in Table XVIII in order to assess the effect of changes in the uncertainties and tolerances.

With a verified empirical fit of the PIF values as a function of the design variables, it will be possible to assess the importance of each variable and to calculate the probability of the PIF value exceeding a fixed quantity. In this way, the PIF values are useful for determining the maximum safe operating power level of AARR.

b. Test Program. The initial fuel power core heat transfer test conditions have been sufficiently established for the core to allow formulation of a heat transfer test program. The range of variables to be covered are listed in Table XIX. These conditions are tentative and are expected to be altered, in response to results obtained as the test program proceeds. Decay-cooling problems will be studied as part of a separate program.

Table XIX. AARR Heat Transfer Test Conditions

Test section heat flux, axially uniform; cosine; skewed.

Channel geometry	right rectangle
Channel spacing, in.	0.030; 0.040; 0.050
Channel width, in.	1.25
Channel exit pressure, psia	665; decreasing pressure
Channel velocity, ft/sec	30; 40; 45; 50; 60
Channel inlet temperature, °F	100; 135; 160
Channel flow	steady; decreased flow
Channel flow direction	downflow, upflow
Channel flow configuration	single channel with and without parallel unheated bypass
Void measurements	yes
Test objectives	flow instability, critical heat flux, critical flow and flow regime

B. Rocket Fuel Test Reactor

The calculations were completed on the study of the use of variable loading of fuel and moderator in the $\text{UO}_2\text{-BeO}$ and U-Zr-ZrH_2 test reactors. The results are presented in Table XX.

Table XX. Data on Variable Loaded Test Reactors

Type	Power Density Ratio		Volume (liter)	Power (MWt)	Power in Uniformly Loaded Test Reactor (MWt)
	$\frac{\text{Max in Test Section}}{\text{Max in Test Reactor}}$	$\frac{\text{Max in Test Section}}{\text{Centerline in Test}}$			
$\text{UO}_2\text{-BeO}$	4.0	1.11	490	740	1050
U-Zr-ZrH_2	4.0	1.04	920	1380	1900-2000

The influence of substituting U^{238} for tungsten in the buffer region between the test section and the test reactor was determined for the BeO type of test reactor. The description of the buffer and the results of the calculations are given in Table XXI.

Table XXI. Influence of Buffer on Rocket Fuel Test Reactor Performance

Buffer		Power Density Ratio		Volume (liter)	Power (MWt)
Material	Thickness (cm)	Maximum Ratio	Maximum Centerline		
W	1	4.0	1.11	490	740
U^{238}	1	4.0	1.09	470	700
W	4	4.0	1.005	825	1640
60% U 40% Na	4	4.0	1.004	705	1160
25% W 45% U^{238} 30% Na	4	4.0	1.004	740	1200

The following conclusions may be drawn:

- (1) A $\text{UO}_2\text{-BeO}$ type of test reactor produces significantly less power than a U-Zr-ZrH_2 test reactor, other factors being equal.
- (2) In general, the power produced in the test reactor varies inversely with the ratio of maximum-to-centerline power densities.
- (3) The performance of the reactor is slightly better with U^{238} in the buffer region than with tungsten.
- (4) The flux spectrum in the test section is more closely simulated with a thicker buffer region, but the power generated in the test reactor is also increased.

C. Magnetohydrodynamics (MHD)

1. MHD Power Generation - Jet Pump Studies

The previous flow rate and efficiency studies with the recirculating jet pump configuration (see Progress Report for January 1964, ANL-6840, p. 87) verified the necessity for effective velocity matching between the vapor discharge from the nozzle and the liquid entering the mixing zone. A possible method for achieving this type of matching lies in staging of the jet pumps.

An arrangement for series operation of two jet pumps was assembled so that a portion of the steam can be taken from the final stage to operate the preceding unit. In this way steam of progressively lower energy content would move to the head of the unit while the liquid velocity will progressively increase toward the outlet of the combination.

A number of runs were tried with a two-stage combination. However, positioning and operation were critical. Several runs had to be performed before pumping was achieved, although not efficiently as yet. Test are continuing.

2. Liquid Metal Pumping

A small model of an MHD loop was constructed as an MHD power demonstration unit for the Geneva Conference. This loop consisted of a series flow NaK circuit with flow supplied by a small DC EM pump. The generator was a plastic duct with electrodes surrounded by a 6,000-gauss permanent magnet. Several tests with the unit showed that a voltage of 50 mV was obtainable, but that the magnet of the generator severely restricted the flow. The generator, however, could be loaded to supply 4 Amp of current.

Comparison has shown that the generator pumps as well as the pump at a power input of approximately 6 W. For this same type of pumping the input to the pump had to be ~240 W. This showed very clearly the effect of the shorting provided by the pump tube. Under these conditions it was possible to pump with the generator by supplying power from a 1.5-V dry cell.

3. Thermionic Arc for MHD Power Generation

The feasibility of applying a thermionic arc to MHD power generation is being considered. This possible process is an alternative method for extraction of electrical power from an MHD generator. It also appears possible to initiate an oscillatory mode of operation to this system so that the electrical power can be removed as an alternating voltage.

4. MHD Duct Flow

A study of the transient response of an electrically conducting fluid to simultaneous changes in the applied magnetic field, duct wall temperature, and the driving pressure gradient has been completed (see Progress Report for October 1963, ANL-6801, p. 69). The effect of five parameters (Hartmann number M , Rayleigh number Ra , thermal and magnetic Prandtl numbers Pr and Pr_m , and energy generation index) upon the flow and heat transfer characteristics in a constant area duct were analyzed.

It was found that severe oscillations in the transient flow rate could occur when the Rayleigh number was large (i.e., when the heat transfer rate between the duct wall and the fluid is large and the fluid has a significant thermal coefficient of expansion). In certain cases, flow reversal was predicted. Similar oscillations were caused by large values of the Hartmann number (i.e., large applied magnetic fields). However, in both cases, these oscillations could be dumped entirely if the thermal or magnetic Prandtl numbers were of the proper magnitude. For very small or very large thermal Prandtl numbers, only a minor transient oscillation in the flow rate occurred; but, when this number was approximately unity, a severe overshoot and oscillation resulted.

The effect of the magnetic Prandtl number on the transient flow rate is even more striking. In Figure 38, the transient flow rate-pressure gradient relationship is shown for the case of $Ra = 10^4$, $M = 100$, $Pr = 0.7$, and zero internal energy generation. The solid line corresponds to the case of $Pr/Pr_m = 1$ (small magnetic diffusivity) and the dashed line to the case of $Pr/Pr_m \gg 1$ (large magnetic diffusivity).

Figure 38. Effect of the Magnetic Prandtl Number on the Transient Flow Rate

When the magnetic diffusivity is large, the applied magnetic field passes through the fluid with little resistance and quickly stabilizes the flow. However, when the magnetic diffusivity is small, there is a considerable time lag between the application of the magnetic field and its complete interaction with the fluid; thus, the oscillations caused by free convective effects cannot be immediately subdued.

Finally, when the fluid generates heat internally, the mean velocity of the fluid is increased (because of free convection effects) and the temperature difference between the fluid and the duct wall is decreased. In

fact, during the transient period, the direction of heat flow may be different from that at steady-state and local temperatures may greatly exceed those calculated at steady-state conditions.

D. Regenerative EMF Cells

1. Bimetallic Cells

a. Sodium-Bismuth System. Studies continued on the development of thermally regenerative emf cells for the direct conversion of heat into electricity. An important part of this program is the assessment of the feasibility of thermal regeneration of the systems under consideration.

A preliminary experiment was carried out to evaluate the thermal regeneration of the sodium-bismuth system. A sample of Na_3Bi (the intermetallic formed in the cathode of the sodium-bismuth emf cell) was heated in a Knudsen effusion cell and the concentrations of the effusing species were measured by means of a mass spectrograph. Only ions of Na^+ , Bi^+ , and Bi_2^+ were found. At 700°K and a residual alloy composition of 45 a/o sodium, the vapor pressure of sodium above the alloy was estimated to be 2.0×10^{-2} mm, whereas the vapor pressure of the bismuth monomer was 8.6×10^{-6} mm. (At this temperature, the concentration of Bi_2^+ is negligible.) During the time required to effuse 73.7% of the sodium originally present in the Na_3Bi phase, only $1.4 \times 10^{-2}\%$ of the bismuth had effused. Despite the preliminary nature of the experiment, the data suggest that thermal regeneration of the sodium-bismuth cell is possible.

b. Spectrophotometric Studies of Li_2Te , Li_3Bi and Na_2Te in Molten Salts. The absorption spectra of Li_2Te and Li_3Bi in molten LiCl-LiF and the spectrum of Na_2Te in molten NaCl-NaI were determined. The absorption spectrum of Li_2Te in the LiCl-LiF salt mixture at 600°C was found to have a single peak at $21,000\text{ cm}^{-1}$. The absorption spectrum of Li_3Bi in LiCl-LiF at 525°C was found to have a single absorption band whose long wavelength edge was located at $16,300\text{ cm}^{-1}$. No peaks were observed in the experiment with sodium telluride. The charge transfer band of Na_2Te in NaCl-NaI at 675°C obscured any absorption peak that might be present above about $15,600\text{ cm}^{-1}$.

c. Solubility of Lithium and Bismuth from a 50 a/o Li-Bi Alloy in Molten LiCl-LiF . The solubility of lithium and bismuth from a 50 a/o lithium-bismuth alloy (presumably dissolving as LiBi and Li_3Bi) in molten LiCl-LiF was determined. The solubility of bismuth increased from 0.53 w/o at 700°C to 1.27 w/o at 1000°C , whereas the solubility of lithium increased from 0.040 w/o to 0.053 w/o when the temperature was increased from 700 to 1000°C . Whereas the total metal solubilities increased with temperature, the ratio of lithium to bismuth decreased with temperature. The Li:Bi ratio was 2.3 at 700°C and 1.3 at 1000°C . The shift in the Li:Bi ratio is expected since the activity of Li_3Bi decreases with increasing temperature.

d. Back-diffusion of Bismuth in Li-Bi Cell. Since it is now known that LiBi and Li_3Bi are soluble as entities in LiCl-LiF (see Progress Report for December 1963, ANL-6810, p. 47), a determination of the back-diffusion of bismuth from a bismuth cathode saturated with Li_3Bi to a lithium anode was undertaken. The experiment was carried out over a period of 51 hr at cell temperatures between 582 and 686°C. The highest concentration of bismuth in the lithium anode was 1.2 m/o.

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Fuel-coolant Chemical Reactions

a. Reaction of Aluminum-Uranium Alloy (SL-1 Material) with Steam. The isothermal reaction of an aluminum-17 w/o uranium alloy (SL-1 composition) with steam is being studied in order to provide a more direct comparison between results of laboratory experiments, data obtained in TREAT experiments, and results of reactor meltdowns. The isothermal rate studies, which are being carried out by means of the levitation method (see Progress Report for January 1964, ANL-6840, p. 92) have been made over the temperature range from 1300 to 1600°C. The duration of the runs varied from 5 to 100 min. A preliminary analysis of the results indicates that the reaction is approximately described by the same cubic rate law which describes the reaction of pure aluminum with steam at temperatures below 1300°C.¹⁸ (Above 1400°C, the reaction of pure aluminum with steam follows a linear rate law.)

b. Laser-beam Heating Applications. Research on the application of laser-beam heating to metal-water reactions continued. Aluminum and zirconium foil samples (1-mm squares) were heated by means of laser beams of various energies while the foils were submerged in water in a sealed quartz cell. The sample weights were about 8×10^{-4} g and 7×10^{-5} g for the zirconium and aluminum samples, respectively. The laser outputs were from 10.4 to 28.7 Joules in the tests with aluminum foils and from 18.8 to 31.6 Joules in the tests with zirconium foils. On melting, the aluminum foils formed 370- μ -diameter spheres and the zirconium foils 630- μ -diameter spheres. The extent of reaction of the metals with water was found to be independent of the laser outputs in both series of experiments. The extent of reaction was about 8% for the aluminum foils and 32% for the zirconium foils. The results of the experiments with zirconium foils are in good agreement with those obtained previously using the condenser-discharge method.¹⁹

c. "Water Hammer" Effect. When a zirconium particle is heated by means of a laser beam while submerged in water in an open tube, the water above the particle rises owing to the rapid generation of a steam-hydrogen bubble. A similar phenomenon was observed when zirconium wires were heated under water by the condenser-discharge method (see Progress Report for November 1963, ANL-6808, p. 73). Under certain conditions, the bubble collapses after a period of about 10 ms and the water column is driven violently toward the bottom of the tube, resulting in the so-called "water hammer."

¹⁸Chemical Engineering Division Summary Report, Oct-Dec 1962, ANL-6648, p. 198.

¹⁹L. Baker, Jr., and L. C. Just, ANL-6548, p. 52.

To ascertain the conditions under which the water-hammer effect could occur, 1-mm and 2-mm squares of zirconium foil were heated by means of a laser beam while submerged under 20 to 160 mm of water contained in open quartz tubes having diameters from 5 to 28 mm. The laser beam outputs were about 30 Joules. High-speed motion picture photography was used to observe the events occurring in the tube.

The geometric requirements were found to be quite critical. The effect was observed only when 2-mm squares of zirconium foils were heated in 5-mm-ID tubes containing 40 mm of water.

d. Studies in TREAT of Oxide-core, Metal-clad Fuel Pins. In the preceding Progress Report (ANL-6840, p. 92), results were given on the extent of the metal-water reaction as a function of energy and on the pressure-rate relationships when stainless steel-clad oxide-core (UO_2) specimens and Zircaloy-clad mixed oxide-core specimens were exposed to TREAT transients while submerged in water. Information concerning the size distribution of particles formed during the experiments is now available. In the case of the stainless steel-clad fuel pins, the Sauter mean particle diameter, d_{sv} ,* was obtained first for the entire residue which included oxide particles and particles of the metal cladding. The Sauter mean diameter of the particles was 26 mils in one run (Run 169) and 10 mils in another (Run 170). The residue was treated with hot nitric acid to dissolve the oxide and the size distribution of the stainless steel particles was determined. The Sauter mean diameter of the stainless steel particles was 59 mils in Run 169 and 82 mils in Run 170. Effective dissolution of the Zircaloy-2-clad mixed-oxide-core specimen (Run 168) could not be achieved because of the presence of insoluble oxides in the core. The Sauter mean particle diameter of the residue (particles from the core and cladding) was 15 mils in the test with the Zircaloy-2-clad fuel specimen.

e. TREAT Experiment with Pre-irradiated Fuel. An additional experiment to determine the effect of pre-irradiation on the extent of metal-water reactions was carried out in TREAT. A pre-irradiated, 20 percent enriched uranium alloy fuel pin (93.5 w/o uranium, 5 w/o zirconium, 1.5 w/o niobium) was used. The fuel pin had been previously irradiated to a 1.5 a/o burnup of the U^{235} present and had been cooled for 2 yr. The fuel pin, submerged in water, was subjected to a TREAT transient which generated 168 MW-sec and resulted in a specific energy input of 150 cal/g to the fuel pin. The fuel pin was completely melted by the TREAT transient. The extent of the metal-water reaction was 10.5%. This result was nearly identical with the results obtained

$$*d_{sv} = \frac{1}{\sum_{i=1}^N W_i/d_i}, \text{ where } d_i = \text{average diameter of } i^{\text{th}} \text{ size group, } W_i = \text{fraction by weight of particles in } i^{\text{th}} \text{ group, and } N \text{ is the number of size groups.}$$

with unirradiated fuel specimens of the same type at a similar energy. The results of this test again indicated, as did the results of a previous test (see Progress Report for December 1963, ANL-6810, p. 53), that pre-irradiation does not have a marked influence on the extent of metal-water reactions.

2. Metal Oxidation-Ignition Studies

a. Effect of Moisture on Oxidation of Plutonium. Previous studies have indicated that the rate of oxidation of plutonium in moist air is markedly higher than its rate of oxidation in dry air (see Progress Report for October 1963, ANL-6801, p. 71). The contribution of the moisture to the oxidation of plutonium was assessed in isothermal oxidation experiments in which plutonium was exposed to an atmosphere of helium containing 20,000 ppm moisture. At 149°C, the oxidation rate of plutonium in this atmosphere was 100 times greater than the oxidation rate in dry oxygen (<5 ppm water vapor) and 10 times greater than the oxidation rate in moist oxygen (<1000 ppm water vapor) at the same temperature.

b. Burning-curve Ignition Studies of Plutonium-containing Alloys. Studies of the ignition temperatures of uranium-30 a/o plutonium ternary alloys by the burning curve method were continued (see Progress Report for December 1963, ANL-6810, p. 50). These studies are being made in an effort to find an alloy of suitable composition for zero-power critical experiments. Alloys having an atomic ratio of uranium-to-plutonium of about 2:1 are desired. However, binary alloys having this composition have a tendency to crumble; and, as a result, work with these alloys presents pyrophoricity hazards.

Previous work (Progress Reports ANL-6808, p. 70, and ANL-6810, p. 50) indicated that ternary alloys containing uranium and plutonium in an atom ratio of about 2:1 with 6 a/o iron or molybdenum added had promising ignition characteristics in dry air. It was also shown that the effect of 20,000 ppm of moisture in the air had a marked effect on the ignition behavior of the iron alloy. Whereas in dry air, the iron alloy did not ignite when heated to 820°C and showed only moderate self-heating in the range of 300 to 450°C, the alloy, in wet air, underwent considerable self-heating and reaction at about 300°C, which resulted in its ignition at about 480°C. The behavior of the 6 a/o molybdenum alloy was essentially the same in wet air as in dry air.

Because the relative humidity at the EBR-II site in Idaho is about 20% (5,000 ppm water vapor), the effect of this concentration of moisture in air on the ignition behavior of the ternary iron alloy was studied. The ignition behavior of the alloy was intermediate between that found in dry air (<5 ppm H₂O) and in air saturated with water at room temperature (20,000 ppm).

c. Aluminum- U_3O_8 Reaction. Additional studies were carried out on the aluminum- U_3O_8 thermite reaction (see Progress Report for November 1963, ANL-6808, p. 71). The re-examination of this reaction was prompted by a publication²⁰ in which it was reported that a thermite reaction occurred in aluminum- U_3O_8 cermet.

In the current studies, a -100 +200 mesh fraction of U_3O_8 was used, whereas in previous studies -400 mesh U_3O_8 was used. The average size of the -100 +200 mesh U_3O_8 particles was found to be 109 μ . (The average size of the -400 mesh U_3O_8 particles used in earlier studies was 12 μ .) The aluminum was -400 mesh powder in both the present and earlier studies. Cold-pressed compacts of U_3O_8 -15 w/o Al and U_3O_8 -25 w/o Al were rapidly inserted into a furnace preheated to 1250°C. The peak temperatures reached by the samples (1300-1400°C) were considerably lower than the peak temperatures reached with samples in which -400 mesh U_3O_8 was used (1600-1800°C). Additional studies are being planned in which pellets prepared from coarse U_3O_8 powder and coarse aluminum powder will be employed.

B. Fast Reactor Safety Studies

Two of the most important design features influencing the safety characteristics of fast power reactors are the large quantity of fuel and the short prompt-neutron lifetime. Therefore, it is possible to postulate a class of "meltdown" accidents, consisting of multistage incidents proceeding as follows: (1) some operating abnormality occurs in which the fuel undergoes failure (or "meltdown"); (2) the material movements produce a more reactive configuration; and (3) a destructive burst of nuclear energy occurs, terminating the accident. The mechanisms involved in fuel failure and material movement are complex, and are influenced by their environment. They are being studied in a program centering on experiments being performed in the TREAT reactor.

1. Integral Sodium Loops

Preparations are under way for performing meltdown experiments on single fuel specimens in flowing sodium using integral loops developed to fit inside the 10-cm-square by 240-cm-long volume of a TREAT fuel element.

The first of the integral sodium loops has reached operational status. The cooling, heating, instrumentation and control circuits of the modified, refitted loop, and the instrument and power circuits of the re-wired control consoles have been tested. All four of the loops have been leak-checked at internal pressures exceeding the maximum pressure anticipated during TREAT experiments.

²⁰J. D. Fleming and J. W. Johnson, *Nucleonics* 21 (5), 84 (1963).

Since the volume in the package loops is limited and high temperatures are to be reached in the planned experiments, it is necessary to measure precisely the amount of sodium in the loop. A controlled-volume, sodium-filling system was designed specifically for the integral loops. This system has been fabricated, fully tested, and is in operation. The first package loop, containing an EBR-II-type fuel element with an internal tantalum-molybdenum thermocouple, is being filled with sodium and an inert gas blanket of helium. The final details for performing the experiment in the TREAT reactor are being arranged.

All components for the remote disassembly apparatus have been received from the shop. A mockup test in the deep pit of Building 308 has been started. Some difficulties have been encountered. For example, interference between the positioner guide rollers and the top portion of the support basket prevented the assembly of the positioner. It has been necessary to fabricate new rollers and modify their supports. Assembly of the equipment is proceeding.

2. Large TREAT Loop

The 4-in. P-K ball valves under repair and modification at DeVar Kinetics are approximately 85% complete at this time. DeVar, who has been analyzing the bodies of the valves to determine the reason for failure during leak rate tests, has been unable to duplicate results on repetitive tests to meet the requirements.

Thermal cycling of the flowmeter magnets has been completed. The large magnet for the primary system remained unchanged. The smaller magnet stabilized after five cycles with a 25% loss during the initial cycling. The magnets are now being installed in the calibration loop at Building 308.

Investigation of the test-section-handling procedures during disassembly indicated that modifications of the shipping casks and handling equipment were necessary. This requirement is necessary for use in the cave facilities of Building 301. The design changes are being made on drawings and the current vendors will be duly advised. Certification of approval for the Shipping Cask has been received from the Bureau of Explosives.

The four vessels for the loop have been completed by the vendor and shipped to the Idaho site. These were code tested and approved by the underwriters insurance for vessel acceptance. All parts were capped or seal welded and the vessels were received at the TREAT facility on February 14.

3. High-temperature Furnace

The tungsten-filament high-temperature furnace which will be used when a general test facility has been assembled is in place. Bus connections to the rectifier power supply have been made, and the hookup of water cooling and blanket-gas lines is being completed. While it is anticipated that the 15-V, 7500-Amp capacity of a single rectifier will be insufficient to reach the 3000°C design capability of the furnace, this power supply is adequate for initial use of the furnace at temperatures of 2000-2200°C. In the first trials of the furnace, the available 0.3-V/tap control in the lowest voltage decade of the rectifier will be used. If this increment of voltage is found to be too coarse, a continuously variable control circuit will have to be incorporated into the first decade of the rectifier supply.

4. Studies of Pressure Transducer Instrumentation

Meltdown experiments with samples submerged in molten sodium require pressure instrumentation capable of providing good transient response under unusual environmental conditions. The frequency-response apparatus for the pressure transducer has been completed and tested. A 0.003-mil annealed copper sheet is used as the diaphragm material in a shock tube. The shock tube generates a steep-fronted pressure pulse of 37.5- μ sec duration and causes the transducer to vibrate at its natural frequency. The response characteristics of the transducer, Type 4-316-CEC, are determined by analysis of the oscillogram of decay. This pressure transducer is expected to have a full amplitude response to pressure pulses as short as 0.6 ms in duration.

VI. PUBLICATIONS

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