ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60440

REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

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Albert V. Crewe, Laboratory Director
Stephen Lawroski, Associate Laboratory Director

<u>Division</u>	<u>Director</u>
Chemical Engineering	R. C. Vogel
Idaho	M. Novick
Metallurgy	F. G. Foote
Reactor Engineering	L. J. Koch
Reactor Physics	R. Avery
Remote Control	R. C. Goertz

Report Coordinated by
R. M. Adams and M. Weber

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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:

November 1963	ANL-6808
December 1963	ANL-6810
January 1964	ANL-6840
February 1964	ANL-6860
March 1964	ANL-6880
April 1964	ANL-6885

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I. BOILING WATER REACTORS

A. BORAX-V

1. Zero-power Experiments

The hot, zero-power measurements made on the peripheral superheater core PSH-1 at conditions up to 600 psig and 489°F were difficult to interpret because the neutron source could not be removed and thus affected the critical control rod positions and the magnitude of the period at low power levels. The period measurements were also affected at higher powers when some boiling occurred.

Figure 1 shows the nine-control-rod-bank differential reactivity worth at various temperatures. The nine-rod-bank worth curve at room temperature, using boric acid to allow the control rods to be withdrawn, is also shown for comparison. At two temperatures, 260 and 485°F, with superheaters flooded, two values of control-rod worth are shown which are quite different in both cases. These are the result of two separate control-rod worth measurements. The differences may have been caused by changes

Figure 1. Nine-control-rod-bank Reactivity Worth at Various Temperatures, Peripheral Superheater Core PSH-1 of BORAX-V

in temperature and pressure, critical control rod position due to the source, or other causes. The hot control-rod worth seems to follow the room temperature curve very closely. However, the hot-rod worth points are probably somewhat low due to the measured critical position being slightly lower than the true critical position because of the source.

The isothermal temperature coefficient in core PSH-1 resulting from these measurements was larger than expected and is shown in Table I, which compares the temperature coefficient and superheater flooding reactivity worths for central superheater core CSH-1 and peripheral superheater core PSH-1. A plot of the temperature coefficient versus temperature is shown in Figure 2. The high, negative temperature coefficient is thought to be due

Table I. Temperature Coefficients and Flooding Worths, Comparison of CSH-1 and PSH-1 Cores of BORAX-V

	<u>PSH-1 Core</u>	<u>CSH-1 Core</u>
Overall Temperature Coefficient from 70 to 489°F, % $\Delta k/k/^\circ F$		
Superheater drained	-0.0128	-0.0085
Superheater flooded	-0.0132	-0.0075
Flooding Worth, % $\Delta k/k$		
Room temperature	1.55	0.86
489°F	1.37	1.27
H ₂ O/UO ₂ Atom Density Ratio of Boiler Fuel Cell at 489°F	4.0	3.5

Figure 2. Isothermal Temperature Coefficient. Superheater Flooded, No Boric Acid, Peripheral Superheater Core PSH-1 of BORAX-V

in part to the placement of the superheater assemblies on the periphery of the core with the attendant leakage effects. The reason for the flooding worth being lower hot than cold in core PSH-1 is not known, but is thought to be partially due to the superheater location. Core CSH-1 contained eight water-filled aluminum flow rods per boiling fuel assembly, while core PSH-1 contained 12 flow rods per assembly.

Flux wire irradiations were made at zero power with the reactor at operating temperature and pressure. Neutron flux distributions with the superheater drained and flooded are shown in Figures 3 and 4, respectively.

Analysis of previously obtained data has resulted in measured values for both radial and axial bucklings. The axial buckling was obtained from a least-squares cosine fit (ID 1620-030) to the symmetrical portion of the curves not influenced by control rods or reflector, and the radial buckling was obtained by drawing a smooth curve through a radial neutron flux plot and fitting both cosine and Bessel functions to this curve. The experimental values are listed in Table II.

Figure 3. Axial Neutron Flux Distributions, Peripheral Superheater Core PSH-1 of BORAX-V

Figure 4 Axial Neutron Flux Distributions, Peripheral Superheater Core PSH-1 of BORAX-V

Table II. Measured Values of Radial and Axial Bucklings,
Peripheral Superheater Core PSH-1 of BORAX-V

Radial				Axial	
Diagonal		Transverse		at Center of Fuel Subassembly	
B^2 (cosine fit) (cm^{-2})	B^2 (J_0 fit) (cm^{-2})	B^2 (cosine fit) (cm^{-2})	B^2 (J_0 fit) (cm^{-2})	B^2 (cm^{-2})	Core Position*
0.00078	0.00171	0.00125	0.00242	0.00362	55
				0.00279	58
				0.00447	57
				0.00343	66
				0.00422	77

* See Figure 1, p. 2, and Figure 3, p. 5, of ANL-6885.

NOTES: Control rods banked at about 9.63 in.; Detector material: U^{235} ;
Temperature: 69°F; Superheater drained.

2. Power Experiments

After completion of the zero-power experiments at operating conditions of 600 psig and 489°F, power operation was started on the core with a peripheral superheater, PSH-1. A top view of this core is shown in Figure 5.

Figure 5. Power Loading Peripheral Superheater Core PSH-1 BORAX-V

The initial approach to full power was made in steps of approximately 2.5 MW. A maximum power of 18.4 MWt and a superheater steam temperature of 820°F at the superheater outlet were achieved on May 20 with all control rods withdrawn and while dissipating power through the turbo-condenser system. Figure 6 is a plot of control rod position versus power during this approach to power. The power coefficient of reactivity was unexpectedly low compared with the high temperature coefficient measured (see Section 1 above).

On May 25, a fast approach to full power was made to minimize xenon build-up by venting steam directly to atmosphere. A reactor power of about 20 MWt and a steam temperature of 832°F were reached with no demineralizer flow. The maximum power of about 22 MWt and superheater exit steam temperature of 890°F were achieved with 40 gpm of reactor water flowing through the reactor water-demineralizer heat exchangers. This bypass flow increased the core inlet subcooling by about 3°F. The feedwater temperature during these maximum power runs was measured as 76°F.

Figure 6. Nine-control-rod-bank Position vs Reactor Power,
Peripheral Superheater Core PSH-1 of BORAX-V

a. Power Measurements. Table III summarizes some of the measurements made during initial power experiments on core PSH-1. The effect of two different control-rod configurations on the boiler-superheater power split and exit steam temperature can be seen. The relation between linear neutron flux and power also changed with control rod configurations, due presumably to control-rod shadowing of the neutron detector. The initiation of 40-gpm flow through the bypass reactor water-demineralizer heat exchangers had a large effect on exit steam temperature.

The large reduction in steam temperatures from the superheater exit to the steam line was caused by the high reactor water levels of 12-14 ft used during initial power operation. The superheated steam flexible hose manifolds in the steam dome were partially immersed in water. Subsequent lowering of the water level has caused an increase in steam-line temperature so that it now approaches the superheater outlet temperature. The leakage

of moderator water into superheater fuel assemblies was remeasured and found to be about 1.3 gph at room temperature, which is not excessive.

Table III. Approach to Full Power, Peripheral Superheater Core PSH-1 of BORAX-IV

Power (MWt)	Linear Neutron Flux (Amps)	9 Control Rod Bank Position (in.)	Central Control Rod Position* (in.)	Feedwater Temperature (°F)	Superheater Entrance Steam Temperature (°F)	Superheater Entrance to 2nd Pass Steam Temperature (°F)	Superheater Exit Steam Temperature (°F)	Steam Line Temperature (°F)	Exit Boiler Void Fraction (Core Position 56) (v/o)	Sub-cooling Temperature (°F)
Steam to Turbine- condenser Experiments										
2.8	0.31×10^{-4}	15.89		95	488	592	690	572	28	1.0
4.8	0.65×10^{-4}	17.00		87	488	497	760	660	37	2.0
4.6	0.67×10^{-4}		7.61	86	488	-	790	669	32	2.0
7.6	0.95×10^{-4}	18.12		109	488	635	777	704	42	2.5
7.0	0.95×10^{-4}		10.23	114	488	650	793	720	39	2.5
10.0	0.13×10^{-3}	19.30		115	488	641	795	720	49	3.5
9.6	0.13×10^{-3}		12.91	116	483	650	803	732	46	3.0
12.2	0.16×10^{-3}	20.39		122	488	640	790	722	54	4.0
12.1	0.16×10^{-3}		15.05	120	488	651	805	731	53	4.0
14.3	0.19×10^{-3}	21.00		116	488	652	812	727	56	4.5
14.2	0.19×10^{-3}		16.95	118	488	651	813	735	56	4.0
18.4	0.25×10^{-3}	25.00		120	488	650	810	720	63	6.5
Steam to Atmosphere Experiments										
20.0 ..	0.29×10^{-3}	25.00		76	487	673	832	760	65	7.0
22.0	0.31×10^{-3}	25.00		76	487	693	890	813	66	10.0

* Remaining eight control rods at 25.00 in.

.. 40 gpm of reactor water flowing through reactor water demineralizer heat exchangers. Returned to feedwater tank.

† Determined from entrance and exit turbine meter readings. Void fraction = $(V_{out} - V_{in}) / V_{out}$, where V_{out} = exit meter equivalent velocity and V_{in} = inlet meter equivalent velocity.

b. Water Chemistry Measurements. During the initial power operation of peripheral superheater core PSH-1, several water chemistry parameters were measured.

Make-up water delivered to the storage tank from the make-up water purification system varied in quality between 15 and 18 megohm-cm. Water delivered to storage at this high quality dropped immediately to 2.5 megohm-cm during low make-up use. During high make-up use, the stored water quality rose as high as 7.5-8.0 megohm-cm.

Quality of feedwater to the reactor varied slowly between limits of 2.0 and 2.6 megohm-cm before steam flow started. At this time, it dropped in about 12 min from 2.3 to 1.8, then rose during a 5-hr period of steam flow until it was up to 7.8. After steam flow stopped, it again dropped to 2.5 megohm-cm in roughly $7\frac{1}{2}$ hours. It slowly dropped to 1.3 megohm-cm during a 2-day shutdown and then began again to improve and rose, with considerable variation, to 2.7 megohm-cm during a 6-hr period of power operation. The slow rises and falls are without obvious cause, except for the marked improvement during steam flow, when the improvement was apparently due to the purified condensate at 10 megohm-cm which then made up the major portion of the feedwater.

Both the reactor water and condensate demineralizers functioned as expected. The condensate demineralizer was used for only about $5\frac{1}{2}$ hr of

the operating period and produced water better than 5 megohm-cm within 20 min of being placed into service at a power level of 2.8 MWt. In 35 more minutes, the effluent was 9-10 megohm-cm, which was maintained during condensate flow.

The reactor water demineralizer was operated only at night except for one 6-hr power run. During this power run, effluent quality was better than 16 megohm-cm.

During the power runs, the pH and resistivity of the reactor water tended to drop as power level increased. Typically, at the start of a run the pH would be 6.5 or above at zero power and 6.1 at 20 MWt. The resistivity would be 2.5 or above at the start, and about 1.2 at 20 MWt. Running the reactor water demineralizer seemed to reduce this trend.

As expected from previous operation, the pH of both saturated and superheated condensed steam was between 5 and 6. None of the values taken seemed to have any trend connected with power level. The pH of the superheated steam had no marked tendency to be lower or higher than the saturated steam.

The resistivity of the condensate from the saturated and superheated steam was quite low, at times reaching values around 500,000 ohm-cm. No values recorded seemed to have any relation to power level, and the superheated steam had no marked tendency to be higher or lower in resistivity than the saturated steam.

Chloride ion concentration of reactor water and condensed saturated steam was monitored. No concentration over 0.04 ppm was observed at any time. The chloride concentration did not increase with increasing power. During the highest power run, the chloride concentration dropped to a value less than the monitor was capable of detecting, i.e., 0.01 ppm or less.

Oxygen concentrations were measured simultaneously on reactor water, saturated steam, and superheated steam. Table IV shows the changes of concentration with power level. Since considerable variation at the lower power levels was observed initially, extreme values are given for the first two days of power operation.

The actual production rate of oxygen in the boiling zone of core PSH-1 is approximated in Figure 7.

Table V gives some of the values of gross specific activity obtained for the reactor water saturated steam and superheated steam. Activity values are arbitrary in counts/min/ml, all determined in identical geometry (well-type crystal). Approximately 15 min was allowed for decay before

counting. The reactor water was sampled in the downcomer region near the mid-point of the vessel. The saturated steam was sampled in the steam dome at the same level as the openings to the first-pass superheaters, and the superheated steam was taken from the main steam line.

Table IV. Oxygen Concentrations, Initial Power Operation of Core PSH-1 of BORAX-V

<u>Date</u>	<u>Time</u>	<u>Power Level (MWt)</u>	<u>Reactor Water (ppm)</u>	<u>Saturated Steam (ppm)</u>	<u>Superheated Steam (ppm)</u>	
May 18	1420-1520	1-2	5.1-0.66	24-27	5-10	
May 19	1320-1328	~1.5	0.2	~33	18.5-26.4	
	1328-1449	4.25	0.2-0.27	31.5-41.5	21.9-36.4	
	1449-1524	5.0	~0.29	24.5-34.5	27.3-43.3	
			<u>Avg. Value</u>	<u>Avg. Value</u>	<u>Avg. Value</u>	
May 20	1120-1145	2.6-2.8	0.20	27.8	28.5	} Steam to Atmosphere
	1145-1310	~4.0	0.25	27.5	27.6	
	1310-1346	~5.0	0.25	23.0	22.9	
	1346-1424	7.6	0.26	21.9	21.2	} Condensate from Turbine-condenser
	1424-1536	9.6	0.26	22.5	22.0	
	1536-1607	12.2	*	22.9	22.5	
	1607-1707	14.3	-	23.0	22.5	
	1707-1750	18.4	-	22.5	21.5	
May 22	1438-1450	3.0	0.20	23.8	45.5	} Steam to Atmosphere
	1450-1504	10.0	0.23	25.0	30.8	
	1504-1508	15.0	0.25	26.5	26.7	
	1508-1513	20.0	0.27	27.0	27.0	
	1513-1520	20.0	0.34	27.5	29.1	
	1520-1530	20.0	0.42	28.5	28.8	
	1530-1540	20.0	0.44	29.5	26.1	
	1540-1546	20.0	0.47	30.5	28.2	
	1546	10.0	0.48	30.5	28.8	

* NOTE: At this time, the oxygen concentration suddenly (in 5 min) rose to 5.3 ppm, dropped sharply (in 10 min) to 1.27 ppm, again rose to 2.8 ppm (in 10 min), then finally began to decrease until at the end of the run it was 0.30 ppm. The thallium oxygen analyzer was checked against the Beckman polarographic analyzer and was reading properly. The reason for this excursion is not known.

Figure 7. Oxygen Production in Boiler, Peripheral Superheater Core PSH-1 of BORAX-V

Table V. Gross Specific Activity, Initial Power
Operation of Core PSH-1 of BORAX-V

Date	Approximate Power Level (Mwt)	Reactor Water (cpm/ml)	Saturated Steam (cpm/ml)	Superheated Steam (cpm/ml)
May 18	1-2	4,700	1,500	1,700
May 19	1	8,000	360	400
	5	39,560	2,080	4,780
May 20	15	268,000	-	9,700
	15	306,000	6,200	9,300
	18	394,000	13,200	13,400
May 22	0.5	7,400	3,500	3,000
	3	18,000	2,560	3,100
	20	78,700	6,800	5,500
May 25	6	12,400	1,500	9,900

The main isotope found in the reactor water and responsible for most of the activity was Na^{24} . A minor isotope of relatively low level not previously found was Co^{60} . Cobalt has been noted before as inactive Co^{59} in emission spectrographic analyses of insoluble solids.

In the superheated steam, a trace of xenon and iodine isotopes was observed. However, the level has not increased.

II. LIQUID-METAL-COOLED REACTORS

A. General Fast Reactor Physics

1. ZPR-III

a. Introduction. The final experiments on Assembly 44, RAPSODIE, were completed this month. In the engineering core mockup, radial and axial traverses were made with fission chambers and perturbation samples (see Progress Report for March 1964, ANL-6880). The four boron carbide rods in the mockup, representing the six control/safety rods of RAPSODIE, were then removed from the assembly, and a Rossi- α measurement was made. Finally, a more exact mockup of the six RAPSODIE boron rods was constructed in the assembly. Worths were obtained for full and half insertion of a RAPSODIE control rod, and insertion of one and then two of the RAPSODIE control rods. With the control rod fully inserted, enriched and depleted uranium foils were irradiated in the rod and core to provide a fine fission foil traverse.

b. Traverse Experiments. The traverse experiments in the four-rod engineering mockup consisted of remotely propelling small fission and reaction chambers and small perturbation samples through a one-half-inch-diameter guide tube installed in the reactor. These traverses were made along the axis of the core and radially near the core midplane. Table VI lists the compositions of the counters and samples used. In addition to the counters listed, traverses were also made with a small BF_3 counter. Figure 8 is a plot of relative count rates versus distance from the core center for the various axial traverses. Figure 9 is a plot of the worth of the axial perturbation traverses. In Figure 8, each count rate has been normalized to unity at the core center. Since the results of the radial traverses are similar, they have not been plotted.

Table VI. Specifications of Traverse Counters and
Traverse Perturbation Samples

<u>Traverse Counters</u>							
Type	Plating Mass (mg)	Composition (a/o)					
		U^{235}	U^{238}	U^{234}	Pu^{239}	Pu^{240}	Pu^{241}
Enriched Uranium	5.1	93	6	1	-	-	-
Depleted Uranium	5.0	80 ppm	100	-	-	-	-
Plutonium	2.04	-	-	-	94.9	4.8	0.3

<u>Perturbation Samples</u>							
Type	Mass (g)	Composition (w/o)					
		U^{235}	U^{238}	U^{234}	Pu^{239}	Pu^{240}	Pu^{241}
Enriched Uranium	21.4	93.0	6.0	1	-	-	-
Natural Uranium	64.4	0.7	99.3	-	-	-	-
Plutonium	24.1	-	-	-	94.5	5.1	0.4

Figure 8. Axial Traverses of Pu, U²³⁵, U²³⁸, Fission Counters and BF₃ Chamber in RAPSODIE Mockup

Figure 9. Axial Traverses of Pu, U²³⁵, U²³⁸, Reactivity Samples in RAPSODIE Mockup

c. Rossi- α . It was preferable to measure Rossi- α in a simpler core; therefore, the four mocked-up RAPSODIE rods were removed from the assembly, along with several liters of core from the core edge. The Rossi- α chambers were then installed and criticality again approached. The value of Rossi- α at delayed critical was measured as $0.77 \times 10^5 \text{ sec}^{-1}$ using H³ counters.

d. Six-rod Engineering Mockup. To construct the six-rod engineering mockup of the RAPSODIE reactor within the available inventory of boron carbide and to simulate realistically the proper positions of the rods, it was necessary to translate the axis of the reactor in the assembly one inch horizontally and one-half inch vertically. The six rods were built into the assembly in the "banked" position, which is with the boron portion of the rod inserted one inch below the top of the core. Criticality was then approached by adding core material at the core edge. Figure 10 shows the core configuration with the rods in the "banked" position, and Table VII lists the compositions of the control and safety rods. The critical volume of the six-rod mockup was 39.06 liters, as compared to 38.85 liters for the four-rod mockup critical volume.

- ZPR III CONTROL ROD
MOCKUP OF RAPSODIE SAFETY ROD (Enriched B_4C)
MOCKUP OF RAPSODIE CONTROL ROD (Natural B_4C)

Figure 10
Critical Core Configuration with Six-rod RAPSODIE Engineering Mockup

Table VI' Composition of RAPSODIE Control and Safety Rod Mockups in Assembly 44

	Control Rods		Safety Rods	
	Active Portion	Stem ^(a)	Active Portion	Stem ^(a)
<u>Dimensions</u>				
Cross-sectional area	21.1 cm ²	21.1 cm ²	21.1 cm ²	21.1 cm ²
Length	45.0 cm	66.0 cm ^(b)	45.0 cm	66.0 cm ^(b)
<u>Composition (g/cm³)^(c)</u>				
B ¹⁰	0.113		1.03	
B ¹¹	0.524		0.116	
C	0.19		0.506	
Na		0.22		0.22 ^(c)
Steel	1.44	5.58	1.52	5.58
Al	0.002			0.1 ^(c)

^(a) The stem refers to the portion of the rod which is above or below the active portion, depending on the degree of insertion of the rod.

^(b) The length of the stem that is below the active portion when the rod is banked.

^(c) In Safety Rods Nos. 1 and 2 the aluminum was not present, in Safety Rods Nos. 3 and 4 the sodium was not present.

The simulated RAPSODIE Control Rod No. 1 was then rebuilt to simulate insertion to the core midplane in the first step, and then to the "fully inserted" position, which is with the boron portion extending one inch below the bottom of the core. The worth of this rod in these positions is given in Table VIII. Fine fission foil traverses were then made through the control rod. These data are being analyzed.

Table VIII. Worth of RAPSODIE Control Rod

<u>Position</u>	<u>Worth (Ih)</u>
All rods banked. Active portions 1 in. below top of core.	0
Bottom of active portion at core midplane.	-69.7
Bottom of active portion 1 in. below bottom of core.	-147.4

With this control rod fully inserted, Safety Rod No. 2 was then rebuilt to simulate an insertion of the active portion to a position one inch below the midplane of the core, and then the fully inserted position. Then, with the control rod and Safety Rod No. 2 fully inserted, Safety Rod No. 1 was rebuilt in the fully inserted position. The worth of these rod positions and the combined worth of the two safety rods are given in Table IX.

Table IX. Worth of RAPSODIE Safety Rods

<u>Configuration</u>	<u>Worth (Ih)</u>
All safety rods banked. One control fully inserted.	0
Safety Rod No. 2, 1 in. below midplane of core.	-496
Safety Rod No. 2, 1 in. below bottom of core.	-939
Safety Rods Nos. 1 and 2, 1 in. below bottom of core.	-1510

With the completion of the rod worth measurements, the Assembly 44, RAPSODIE, experiments were terminated.

2. ZPR-VI

a. General. Sodium-void coefficient studies were made throughout ZPR-VI Assembly No. 2, which has been described in previous Progress Reports. Each drawer in the core consists of two $\frac{1}{16}$ -in. columns of enriched uranium, four $\frac{1}{4}$ -in. columns of canned sodium, three $\frac{1}{8}$ -in. columns of graphite, and four $\frac{1}{8}$ -in. columns of depleted uranium. The core half-height is 18 in. Sodium-versus-void coefficients were made by substituting either empty stainless steel cans or stainless steel frames having the same weight as an empty can in various sectors of the assembly for the four columns of sodium-filled stainless steel cans in each drawer. The sectors where these replacements were made are shown in dark outline in Figures 11A through 11J. The top view of each figure indicates the vertical cross section of the assembly; the bottom view of each figure indicates the

Figure 11A-J. Schematic of Core Loading for Performing Sodium-removal Experiments

horizontal cross-section where the replacement experiments were carried out. The change in reactivity was determined with the use of a calibrated control rod. The experimental results, with corrections and theoretical calculations based on diffusion theory are shown for many of the experiments on the data tables (Tables X-XIX) for the corresponding figures.

Table X. Data for Figure 11A

Experimental Results for 1.21 kg of Sodium Removed	Rod No. 8	-2.07	} Ih
	Other Rods	+0.14	
	Table	-0.06	
		-2.0	Ih
Experimental Sodium Coefficient		-1.6	} Ih/kg
Calculated Sodium Coefficient (Set 199)		-1.47	
Calculated Sodium Coefficient (Set 635)		+0.32	

Table XI. Data for Figure 11B

Experimental Results for 5.69 kg of Sodium Removed	Rod No. 8	+18.12	} Ih	Rod No. 8	-10.04	} Ih
	Rod No. 9	-30.62		Rod No. 9	-	
	Other Rods	-0.04		Other Rods	0.00	
	Table	-0.20		Table	+0.13	
	SS (Idaho Cans)	+1.00		SS (Frames)	-1.97	
		-11.7	Ih		-11.9	Ih
Experimental Sodium Coefficient		-2.07	} Ih/kg			
Calculated Sodium Coefficient (Set 199)		-2.05				
Calculated Sodium Coefficient (Set 635)		-0.44				
Calculated Sodium Coefficient (Set 201)		-0.64				

Table XII. Data for Figure 11C

		Whole Region (15.04 kg of Sodium)	Half Region* (7.52 kg of Sodium)
Experimental Results	Rod No. 8	-46.75	+22.63
	Other Rods	+0.07	+0.04
	Table	-0.13	+0.25
	SS (Idaho Cans)	+1.83	-0.92
		-45.0 Ih	+22.0 Ih
Experimental Sodium Coefficient		-2.99	-2.93 Ih/kg
Calculated Sodium Coefficient (Set 199)		-2.80	
Calculated Sodium Coefficient (Set 635)		-1.30	

*This experiment consisted in the replacement of sodium for void in the movable half when the whole region was already voided.

Table XIII. Data for Figure 11D

Experimental Results for 7.53 kg of Sodium Removed	Rod No. 8	-49.00	} Ih
	Other Rods	0.00	
	Table	+0.38	
	SS (Idaho Cans)	+0.96	
		-47.7	Ih
Experimental Sodium Coefficient		-6.34 Ih/kg	

Table XIV. Data for Figure 11E

Experimental Results for 17.22 kg of Sodium Removed	Rod No. 8	-84.70	} Ih
	Other Rods	-0.01	
	Table	-0.13	
	SS (Idaho Cans)	-0.55	
		-84.3	Ih
Experimental Sodium Coefficient		-4.89 Ih/kg	

Table XV. Data for Figure 11F

Experimental Results for 3.79 kg of Sodium Removed	Rod No. 8	-17.0 Ih
	No correction made	
		-17.0 Ih
Experimental Sodium Coefficient		-4.5 Ih/kg

Table XVI. Data for Figure 11G

	Region of Measurement	Inhours		
		A	B	C
Experimental Results for 1.90 kg of Sodium Removed	Rod No. 8	-3.60	-7.71	-14.05
	Other Rods	-0.08	+0.06	+0.00
	Table	-0.06	+0.00	-0.19
		-3.6 Ih	-7.7 Ih	-14.2 Ih
Experimental Sodium Coefficient		-1.9 Ih/kg	-4.0 Ih/kg	-7.5 Ih/kg

Table XVII. Data for Figure 11H

	Region of Measurement	Inhours		
		A	B	C
Experimental Results for 1.99 kg of Sodium Removed	Rod No. 8	-7.97	-7.69	-9.64
	Other Rods	-0.03	-0.14	-0.09
	Table	+0.13	+0.25	-0.19
	C R C *	-0.95	-0.90	-0.97
		-8.8 Ih	-8.5 Ih	-10.9 Ih
Experimental Sodium Coefficient		-4.4 Ih/kg	-4.3 Ih/kg	-5.5 Ih/kg

*C R C. is the control rods' contribution obtained from extrapolation. The two rods were ideally part of these half rings, but were excluded because their loading and unloading were too time-consuming.

Table XVIII. Data for Figure 11I

	Region of Measurement	Inhours			
		A	B	C*	
Experimental	Rod No. 8	-34.52	-31.96	-14.07	-14.52
Results for	Other Rods	-0.06	-0.03	+0.04	+0.00
5.79 kg of	Table	+0.00	+0.13	-0.06	+0.06
Sodium Removed	SS (Idaho Cans)	+0.00	-0.13	-0.28	-0.28
		-34.6 Ih	-32.0 Ih	-14.4 Ih	-14.7 Ih
Experimental Sodium Coefficient		-5.96 Ih/kg	-5.52 Ih/kg	-5.02 Ih/kg	

*The void effect in this ring was measured in two steps. First the void was made in the bottom half (-14.4 Ih); then the top half was voided (-14.7 Ih); -5.02 Ih/kg is the resulting coefficient for the whole region voided (-29.1 Ih)

Table XIX. Data for Figure 11J

	Region of Measurement	Inhours		
		A	B	C
Experimental	Rod No. 8	-30.48	-21.95	-13.98
Results for	Other Rods	-0.07	+0.09	-0.02
4.20 kg of	Table	+0.19	-0.19	+0.00
Sodium Removed	SS (Idaho Cans)	-0.27	-0.25	-0.25
		-30.5 Ih	-22.3 Ih	-14.2 Ih
Experimental Sodium Coefficient		-7.26 Ih/kg	-5.31 Ih/kg	-3.39 Ih/kg

Central reactivity coefficients of various materials were measured at the center of Assembly No. 2. These measurements were made by replacing a 2-in. x 2-in. x 1-in. void at the interface of the two halves at the center of the core with different materials. With the exception of the fissile materials, all the reactivity measurements were made using a remotely operated sample changer. The sample-changer drawer replaced the normal full drawer located at S2323 at the center of the stationary half of the assembly. The materials used and the reactivity coefficients obtained are listed in Table XX.

Table XX. Central Reactivity Coefficients

Material	Material Weight (gm)	Ih/kg	Material	Material Weight (gm)	Ih/kg	Material	Material Weight (gm)	Ih/kg
U ²³⁵	272.95	+104.4 ± 1.5	W (No. 4)	211.44	-8.25 ± 0.27	Fe	488	-2.87 ± 0.10
Pu	123.46	+158.7 ± 4.1	W ¹⁸⁶ (No. 6)	209.37	-6.96 ± 0.27	Fe ₂ O ₃	137.3	-0.13 ± 0.40
Pu	92.97	+158.1 ± 5.4	W ¹⁸⁴ O ₃ (No. 7)	163.59	-5.91 ± 0.34	UO ₂	152.3	-4.26 ± 0.36
W ₂ O ₃ (No. 0)	177.08	-2.72 ± 0.32	D.U.	1154	-6.24 ± 0.04	Cr	220.73	-3.13 ± 0.25
W (No. 5)	224.47	-11.4 ± 0.25	Al	166.9	-1.71 ± 0.44	Nb	481.20	-16.9 ± 0.11
W	1052	-10.9 ± 0.05	Al ₂ O ₃	155.2	+2.50 ± 0.32	B ¹⁰	33.30	-1843.0 ± 7.0
W (No. 1)	257.85	-12.2 ± 0.22	SS-304	499.6	-3.45 ± 0.1		29.07 (B ¹⁰)	
W (No. 2)	173.74	-11.0 ± 0.32	Au	1183.6	-18.4 ± 0.04			
W (No. 3)	237.14	-10.1 ± 0.24	C	103	+16.9 ± 0.5			

The tolerances shown for each sample in Table XX represent the precision in the reproduction of the change in control-rod position when a reactivity measurement was made. The measured reactivity is based on a value of 2.5 lh/cm for the control rod in the range of interest. The tolerances quoted for each sample do not reflect any uncertainties in the control-rod calibration.

The isotopic compositions of the various tungsten samples are listed in Table XXI.

Table XXI. Isotopic Composition of Tungsten Samples

Material	Total Weight (gm)	Isotopic Composition (%)				
		W ¹⁸²	W ¹⁸³	W ¹⁸⁴	W ¹⁸⁶	
WO ₃ (No. 0)	177.08	26.4	14.4	30.6	28.4	Normal W
W (No. 5)	224.47	26.4	14.4	30.6	28.4	
W (No. 1)	257.85	36.9	17.7	31.5	13.9	
W (No. 2)	173.74	24.1	14.7	34.2	26.9	
W (No. 3)	237.14	12.5	9.8	30.8	46.8	
W (No. 4)	211.44	5.8	5.7	23.3	65.1	
W ¹⁸⁶ (No. 6)	209.37	1.6	2.1	11.6	84.7	
W ¹⁸⁴ O ₃ (No. 7)	163.59	1.91	1.87	94.3	1.91	

b. Fine Structure of U²³⁵ and U²³⁸ Fission Rates - Assembly No. 2, ZPR-VI. The distribution of the U²³⁵ and U²³⁸ fission rates across the heterogeneous structure of a core drawer was measured by irradiating a set of enriched and highly depleted uranium foils. Pairs of these foils (0.365 in. in diameter, 0.005 in. thick) were placed between the mockup plates of drawer S2325 at a distance of 4 in. from the interface. Gamma activities above 412 keV were measured relative to the gamma activity of enriched and depleted foils. The foils were irradiated during the same run at the center of the core between the sensitive areas of Kirn-type U²³⁵ and U²³⁸ fission counters, thus providing a calibration of foil activities in terms of absolute fission rates.

The measured traverses (Figure 12) show a smooth variation of the U²³⁵ fission rate, similar to the macroscopic flux distribution, and a pronounced, fine structure in the U²³⁸ fission rate with maxima at the fuel columns. The resulting local fission ratio, $\sigma^{28}/\sigma^{25}(z)$, varies within +10% and -7% about the ratio of the volume-averaged fission rates, $\bar{\sigma}^{28}/\bar{\sigma}^{25} = 0.0359$, which represents the fission ratio in the volume-averaged neutron spectrum. The fission ratio, $\sigma^{28}/\sigma^{25} = 0.0346$, measured with the gas-flow counters is 3.8% below this average, showing that the spectrum at the detector position is slightly softer than the volume-averaged spectrum.

Figure 12
Fine Structure of Fission
Rates in Core Drawer

3. ZPR-IX

Previous progress reports on the aluminum-reflected tungsten-based fast-reactor experimental program were reported in the Progress Reports for February and March 1964, ANL-6860 and ANL-6880. The present report summarizes the first three assemblies studied in the ZPR-IX program (with particular emphasis on Assembly No. 3) in which the primary goal was to establish a confidence level for the available tungsten cross-section sets, particularly the high-energy, inelastic-scattering constants. All cores were made up of parallel $\frac{1}{8}$ -in. thick columns of fuel and diluent loaded into 2-in. x 2-in. aluminum drawers. When assembled in the aluminum matrix on 5.54-cm centers, the drawers form right cylinders with L/D ratio between 0.8 and 1.0. For each case, central reactivity worths were determined for a number of "standard" samples.

The ZPR-IX Assembly No. 1 core composition was identical to that of ZPR-VI Assembly No. 1, except that a 30-cm aluminum reflector was substituted for the depleted uranium reflector. Both cores were studied extensively and had a 7:1 volume ratio of depleted-to-enriched uranium. In succeeding

assemblies, columns of depleted uranium were systematically replaced by tungsten. A typical drawer loading and a cross-section map of Assembly No. 3 is shown in Figures 13 and 14, respectively.

Figure 13

Schematic View of Composition of a Typical ZPR-IX Drawer

Figure 14

Cross Section of Assembly No. 3 on ZPR-IX, Showing Location of Control Rods (Stationary Half) and Core and Reflector Boundaries. Movable half control rods are not symmetrical with those shown.

Table XXII indicates the relative volume fractions of enriched and depleted uranium and tungsten for each of the three assemblies studied, as well as for the reference assembly. Volumes, critical masses, and dimensions are also listed. The experimental critical masses have been corrected for the excess reactivity in terms of peripheral fuel worth. They do not include shape factors or factors for heterogeneity effects, central gap worth, or the irregularity of the cylindrical boundary.

a. Central Worths. Central reactivity worths of "standard" samples of rhenium and tungsten metal and of 94.5% enriched B^{10} powder, encased in stainless steel containers, were measured in each of the ZPR-IX assemblies. The results are listed in Table XXIII. Measurements were

made relative to a calibrated control rod and required the reactor to be shut down and the tables separated in order to change samples. An uncertainty of slightly less than one lh is associated with re-establishment of the critical rod and table positions under these conditions. All measurements are with respect to a 2-in. x 2-in. x 1-in. void in the center of the core and have been corrected for clad material, where appropriate.

Table XXII. Physical Parameters ZPR-VI Reference and ZPR-IX Assemblies

	Reference ZPR-VI Assembly 1	ZPR-IX		
		Assembly 1	Assembly 2	Assembly 3
Crit. Mass (kg U^{235})	234.8	287.5	382.6	493.0
W: U^{238} : U^{235} :vol ratio	0:14:2	0:14:2	4:10:2	7:7:2
Length/Radius, cm	50.8/29.0	50.8/31.4	60.96/34.12	71.4/35.5
Vol. cm^3	159	158	224	283
Atom Density ($\times 10^{24}$)				
U^{235}	0.00426	0.00451	0.00451	0.00447
U^{238}	0.0339	0.03380	0.02430	0.01684
W	0	0	0.01227	0.02138
Al	0	0.00665	0.00680	0.00681
Fe	0.006719	0.00052	0.00022	0.00020
Cr	0.0009	0	0	0

Table XXIII. Central Reactivity Worths

	Reference ZPR-VI Assembly 1	ZPR-IX					
		Assembly 1		Assembly 2		Assembly 3	
		Wt (kg)	ρ/kg	Wt (kg)	ρ/kg	Wt (kg)	ρ/kg
U^{235}	+253.4 ρ/kg	0.268	+240	0.1364	+189	0.1364	+162.4
U^{238}	-12.7 ρ/kg	1.198	-10.9	0.8948	-8.96	0.531	-6.33
W	-24.6 ρ/kg	1.052	-22.6	1.052	-17.6	1.162	-14.8
Re	-64.7 ρ/kg	1.266	-60.0	1.266	-49.3	1.270	-44.3
B^{10}	-2908 ρ/kg	0.0292	-2840	0.0292	-2397	0.0292	-2110
Graphite	-27.8 ρ/kg	0.103	-37.3			0.103	-37.9
Au		1.184	-33.7	1.184	-27.4	1.184	-23.4

b. Hydrogen Worth. A limited number of studies involving hydrogen worths were made in Assembly No. 2. Various combinations of epoxy and B_4C impregnated epoxy plates, each $\frac{1}{8}$ in. x 2 in. x 2 in., were measured separately and intermixed. These studies were repeated in Assembly No. 3. Results of these studies are shown in Table XXIV.

The change in sign for the pure epoxy between the two cores remains unexplained. The work with epoxy is somewhat qualitative in nature since an accurate hydrogen-carbon composition of the material is not known.

Table XXIV Epoxy Worth Measurements

Sample	Epoxy Wt (gm)	B ₄ C Wt (gm)	Meas. Worth, Assembly 2	Meas. Worth, Assembly 3
8 Plates Epoxy	81.1883	0	+9.75	-4.46
4 Plates Epoxy	39.699	0	+0.33	-4.31
1 Impregnated Plate	9.755	14.62	-10.21	-10.55
2 Epoxy + 1 Impreg. + 2 Epoxy	51.244	14.62	-27.56	-27.41

More extensive studies were made in Assembly No. 3 using Lucite and polyethylene of the following compositions:

	H, w/o	C, w/o	O, w/o
Lucite	8.05	59.99	31.96
Polyethylene	14.37	85.63	0

The central worth of hydrogen was determined as a function of the number of hydrogen atoms present using 1-in. x 2-in. x 0.02-in. polyethylene and 2-in. x 2-in. x 0.25-in. Lucite pieces, singly and together. Since the composition of each material is accurately known, correction can be made for the negative reactivity contribution of carbon, assuming this is linear in the weight range studied. The samples and individual results are given in Table XXV. The carbon correction was calculated, using the central worth of 37.9 lh/kg of graphite and subtracted from the sample worths.

Table XXV. Central Worth of Hydrogen

Material	Weight (gm)	Sample Worths, lh (rel. to air void)	Hydrogen Worth (lh/kg)
Polyethylene	4.74	-2.04	-2770
Lucite	17.86	-3.32	-2020
Lucite	35.68	-4.62	-1330
Lucite	52.26	-4.80	-858
Lucite	71.05	-4.35	-477
{ Lucite and	35.68 }	2.66	-123
{ Polyethylene	29.96 }		
Polyethylene	59.92	1.34	-49

c. Peripheral Material Worths for Assembly No. 3. To obtain preliminary estimates of the effects of hydrogen in the reflector, several reactivity measurements were made with 0.020-in.-thick slabs of polyethylene, both normal and parallel to the fuel columns, at the core-reflector interface, and 5.55 cm into the reflector. In addition, a number of reflector materials were tested. These results are summarized in Table XXVI.

Table XXVI. Reflector Material Analyses

Material	Location*	Weight	Reac. Effect (lh)
4-in. x 15-in. x 0.020-in. slab polyethylene	Parallel	19.4 gm	-1.33
8-in. x 15-in. x 0.020-in. slab polyethylene	Normal	38.7 gm	-1.79
26-in. x 15-in. x 0.020-in. slab polyethylene	Normal	129.0	-6.48
26-in. x 15-in. x 0.020-in. slab polyethylene	5.55 cm ⁺	129.0	-1.02
10-in. x 15-in. x $\frac{1}{8}$ -in. Al	Normal	458.0 gm	+1.50 lh/kg ^{††}
10-in. x 15-in. x $\frac{1}{8}$ -in. Graphite	Normal	497.0 gm	+4.63 lh/kg ^{††}
10-in. x 15-in. x $\frac{1}{8}$ -in. W	Normal	5.47 kg	+0.22 lh/kg ^{††}
25-in. x 15-in. x $\frac{1}{4}$ -in. W	Normal	28.5 kg	+0.18 lh/kg ^{††}
10 drawers at periphery - aluminum		13.7 kg	+3.23 lh/kg ^{††}
10 drawers at periphery - Al ₂ O ₃		21.9 kg	+4.57 lh/kg ^{††}
10 drawers at periphery - graphite		14.7 kg	+6.66 lh/kg ^{††}
10 drawers at periphery - oxygen**		10.3 kg	+6.09 lh/kg ^{††}

*Parallel slabs were located on the sides of the core. Normal slabs were located at the top of the core.

**By inference from aluminum and Al₂O₃ studies.

[†]Into reflector, normal to fuel column.

^{††}Relative to void.

d. Fission Ratios. The measured U²³⁸ to U²³⁵ fission ratio for Assembly No. 3 was 0.0330 at the core center, and it was quite constant in the radial direction to within 1 cm of the core-reflector interface. This was in good agreement with the calculated value of 0.035. In addition, Pu²³⁹ and Np²³⁷ fission traverses and a B¹⁰ reaction rate traverse were made through the core and into the reflector. These data are shown in Figure 15.

Figure 15

Fission and Reaction Rate Traverses in ZPR-IX, Assembly No. 3, and U²³⁸ to U²³⁵ Fission Ratio Plot. U²³⁸ and U²³⁵ data were obtained by foil activations, others by solid state detector traverses.

e. Rossi-alpha. Previous measurements of Rossi-alpha ($= \beta/\ell$) in Assembly No. 2 showed a strong dependence on the channel width used in making the measurement as well as the reactor power level. An extended series of measurements in which both variables were studied has been completed and will be reported as a separate matter. For comparison, only the data measured with 0.8- μ sec channel widths and essentially zero power are reported in Table XXVII.

Table XXVII. Kinetic Parameters

	ZPR-VI	ZPR-IX		
	Assembly No. 1	Assembly No. 1	Assembly No. 2	Assembly No. 3
Meas. Rossi- α (sec^{-1})	$(10.0 \pm 0.5) \times 10^4$	$(7.8 \pm 0.3) \times 10^4$	$(7.3 \pm 0.7) \times 10^4$	$(6.90 \pm 0.23) \times 10^4$
β_{eff} (calc.)	0.0073	0.00728	0.00723	0.00708
ℓ (nanoseconds)	73	93	99	103
ℓ (calculated)	63	61	74	82

4. Instrumentation Development

Work on the fast neutron spectrometer has been centered on the machine calculations for "unfolding" the experimental data to give the neutron spectrum. The results have been troubled by instabilities of the calculated results. Work is continuing.

An attempt to check the fissile contents of some ZPR-III fission foils was made by comparing their fission rates in a thermal flux. In this experiment, U^{233} , U^{235} , and Pu^{239} foils were, in turn, mounted in an aluminum-walled fission chamber which was inserted in the thermal column of AFSR. A multichannel analyzer was used to record the fission-chamber count rates. These results, although preliminary, are encouraging in that they tend to confirm that the relative amounts of fissile materials on the foils are consistent with the weights deposited by electroplating.

Intercalibration of a combined $\text{U}^{235}/\text{B}^{10}$ counter by irradiation in a thermal flux was also initiated.

B. General Fast Reactor Fuel Development

1. Metallic Fuels

Uranium-plutonium-base alloys are being investigated for use as fertile-fissile fuel materials in fast breeder reactors because many combinations of the base elements, which are readily available, have good neutron yields and breeding ratios. Alloying additions appear necessary, however, to improve the stability of the uranium-plutonium base in a fast-reactor environment and the compatibility of the fertile-fissile alloy with jacketing materials. Fizzium (Fz), which is a relatively high-zirconium

combination of desirable fission-product metals retained in the fuel on re-processing, has good alloying possibilities. The properties and behavior of uranium-plutonium-fizzium (U-Pu-Fz) alloys, as well as various combinations of the constituent elements, are therefore being investigated.

a. Compatibility of U-Pu-Fz with V-20 w/o Ti Alloy. To help in the selection of fuel and cladding materials for future fast reactors, the

Figure 16. Variation of the Penetration Coefficient with Temperature for V-20 w/o Ti vs. U-10 w/o Pu-10 w/o Fz

by the slope of log-penetration-versus-log-time plot for this combination. The equation describing Figure 16 is

$$K = 0.17 \exp(-74,300/RT).$$

This equation should, however, only be used as a convenience to calculate K at any given temperature and not as a calculation of the activation energy of the process.

compatibility of U-Pu-Fz alloys with potential cladding materials has been studied. On the basis of results of comparative compatibility tests and of comparative sodium-corrosion tests, V-20 w/o Ti, which is fabricable, has been singled out as a promising cladding material for U-10 w/o Pu-10 w/o Fz.

The emphasis on V-20 w/o Ti as the cladding for U-10 w/o Pu-10 w/o Fz fuel requires an expansion of knowledge of their compatibility. Figure 16 is a plot of the penetration coefficient K , versus the reciprocal of the test temperature for a series V-20 w/o Ti versus U-10 w/o Pu-10 w/o Fz diffusion couples annealed from 550 to 700°C, for times from 7 to 104 days. The penetration coefficients are given by the equation

$$X^{3.5} = Kt,$$

where X is the penetration into the cladding in centimeters, t the time for penetration in seconds, and K the penetration coefficient in cm^{3.5}/sec.

The exponent 3.5 was determined

b. High-temperature Irradiations of Metallic Fast Reactor Fuels.

Important aspects of the program for the development of fuels for fast breeder power reactors are the determination of the irradiation stability

of specific fuel compositions and the development of means of controlling irradiation growth. In addition to the uranium-plutonium metallic fuels, thorium-uranium and thorium-plutonium fuels are also being studied.

Five instrumented capsules containing fuels of various compositions are being irradiated in the CP-5 reactor. The capsules contain specimens designed for evaluating the restraint characteristics (including the effect of wall thickness) of various cladding materials that were selected mostly on the basis of high-temperature strength and availability.*

Capsule CP-32 contains specimens fueled with U-20 w/o Pu-10 w/o Fz alloy. The cladding material is Nb-1 w/o Zr, 3.96 mm ID, and the cladding thickness is 0.38 or 0.64 mm. The present maximum cladding temperature is 500°C. The specimens have been irradiated for a calculated 3.3 a/o burnup.

The specimens in capsule CP-33 have fuel concentrations that are either 10 w/o or 15 w/o plutonium with 10% fissionium, the rest uranium. The Nb-1 w/o Zr clad ID is 3.96 mm, and the clad thickness is 0.38, 0.51, or 0.63 mm. The present maximum clad temperature is 430°C. The specimens have been irradiated for a calculated fuel burnup of 3.1 a/o burnup for the 15 w/o plutonium, and 2.6 a/o burnup for the 10 w/o plutonium.

Capsules CP-32 and CP-33 have recently been examined non-destructively by neutron radiography, and all specimens appeared to be intact. The instrumenting thermocouples monitoring the specimen temperatures have not indicated any deformation of the specimens. The irradiation of these two capsules will be continued until there is evidence that one or more of the claddings have failed.

Capsule CP-35 contains specimens fueled with U-20 w/o Pu-10 w/o Fz alloy. Two fuel pins have a 3.33-mm diameter instead of the standard 3.66 mm. Four fuel pins have a 4.4-mm axial hole. The cladding material is Nb-1 w/o Zr alloy, having an ID of 3.96 mm and a wall thickness of 0.23 mm. The present maximum clad temperature is 555°C. The specimens have been irradiated for a calculated 1.8 a/o burnup.

Capsule CP-36 was removed from the reactor on April 27 when a thermocouple indicated a cladding failure. The capsule contained specimens fueled with U-15 w/o Pu-10 w/o Fz alloy and clad in Hastelloy X tubing lined with tungsten. The specimens were being irradiated at a maximum cladding temperature of 680°C at time of failure and had attained 1.2 a/o fuel burnup. This capsule is being disassembled and the specimens examined destructively.

*When the tests were started, Nb-1 w/o Zr appeared to be the most promising of the refractory metals then available. The vanadium-titanium alloys were not then readily available and are still undergoing process development.

Capsule CP-37 is similar to capsule CP-36, differing only in cladding, which in this capsule is 304 stainless steel with a tungsten liner. The present maximum clad temperature is 575°C, and the calculated burnup is 1.1 a/o.

Capsule CP-42 contains specimens fueled with Th-35 w/o Pu, Th-20 w/o U-20 w/o Pu and Th-20 w/o U. The cladding material is Nb-1 w/o Zr of 3.96- and 6.52-mm ID dimensions. The maximum fuel temperature is being maintained at 740°C. The present calculated maximum burnup is 0.3 a/o.

c. Irradiation of EBR-II Prototype U-Pu-Fz Alloy Fuel Rods. In another series of tests, instrumented capsules containing fuel rods with plutonium alloy fuel are being irradiated in the MTR. The fuel rods are structurally similar to those in EBR-II. The capsule has an annulus containing a flowing mixture of helium and nitrogen. The gas composition can be adjusted to provide a fairly wide range of temperature control within the capsule. The fuel rods are submerged in NaK as a heat-transfer medium. A tantalum cup around the fuel rods constitutes a barrier against reaction between the stainless steel of the capsule wall and the plutonium of the fuel in the event that a fuel rod should rupture. At least six thermocouples in the capsule provide temperature indications adjacent to the surface of the cladding on the fuel.

At present, there is one capsule containing a single fuel rod, and two capsules with three fuel rods each, in the reactor. A third capsule with three fuel rods is waiting to be put into the reactor. The maximum estimated burnup achieved thus far is 1.6 a/o. Calculated maximum fuel temperatures are from 550° to 600°C. There have been no indications of ruptured fuel rods made evident by erratic thermocouple readings. The capsules and the fuel rods are identified in Table XXVIII. Variables in these experiments are fuel composition, cladding composition and condition, cladding wall thickness, and void volume. The capsules will remain in the reactor until at least an estimated 2 a/o burnup is achieved or until there is evidence of a ruptured fuel rod.

Table XXVIII. Identification of Capsules and EBR-II Prototype U-Pu-Fz Alloy Fuel Rods

Capsule No.	Date of Insertion	Spec. No.	Fuel Composition*	Cladding**	Cladding, ID, mm	Cladding Thickness, mm	Length of Rod, cm	Length of Fuel Pin, cm	Maximum Fuel Temp., °C	Est. a/o Burnup
ANL-55-5	8-19-63	C-102	U-20 Pu-10 Fs	Nb-1 Zr (SR)	3.96	0.23	46.8	36.1	555	1.6
ANL-55-6	4-15-63	C-94	U-20 Pu-10 Fs	Nb-1 Zr (SR)	3.96	0.23	46.8	36.1	550	1.6
		C-95	U-20 Pu-10 Fs	Nb-1 Zr (SR)	3.96	0.23	46.8	36.1	550	1.6
		C-96	U-20 Pu-10 Fs	Nb-1 Zr (SR)	3.96	0.23	46.8	36.1	550	1.6
ANL-55-7	2-3-64	CB01	U-15 Pu-10 Fz	Nb-1 Zr (SR)	3.96	0.38	46.0	33.0	600	0.35
		EA01	U-15 Pu-10 Fz	Vanadium (A)	3.96	0.38	46.0	33.0	600	0.35
		LA01	U-15 Pu-10 Fz	Hast. X †	3.88	0.27 †	41.7	27.9	600	0.35
ANL-55-8	-	CE03	U-15 Pu-10 Fz	Nb-1 Zr (A)	3.96	0.38	46.1	33.0	-	None
		CK01	U-10 Pu-10 Fz	Nb-1 Zr (A)	3.96	0.38	46.1	33.0	-	None
		EC01	U-15 Pu-10 Fz	Vanadium (A)	3.96	0.38	46.1	33.0	-	None

*20% Pu alloy, uranium not enriched; 15% Pu alloy, uranium enriched 7.7%; 10% Pu alloy, uranium enriched 15.5%.

** (A) Annealed; (SR) Stress Relieved.

† 0.23 mm of annealed Hastelloy X with a tungsten liner 0.04 mm thick.

d. Transient Irradiations of EBR-II Prototype Thorium-Uranium Alloy Fuel Element. As mentioned in the April 1964 Progress Report (ANL-6885, p. 41), neutron irradiation of thorium causes a complex series of nuclear reactions leading to the production of uranium-233, a valuable nuclear fuel. Although these reactions are well-known, more information is required about the properties and reactor behavior of thorium and its alloys.

One question that arises concerns the behavior on thermal excursions of thorium-uranium fuels in which the uranium content greatly exceeds its solid-solubility limit, which is only about 2 w/o. Coalescence of excess uranium existing as a dispersed phase in such alloys was known to occur on prolonged holding at high temperatures; but little was known about the effect of short-time, radiation-induced heating of such alloys to the vicinity of the uranium melting temperature.

Six Th-22 w/o U specimens were therefore subjected to elevated-temperature transients in the TREAT reactor. The sodium-bonded specimens, which were jacketed with 0.38-mm-thick Nb-1 w/o Zr, or Type 304 stainless steel tubing, had no previous irradiation history. Measured cladding temperatures ranged from 735° to 1435°C. The posttransient examination revealed that the integrity of the jacketing on all six specimens had been maintained. There were no dimensional or volume changes on jacketed specimens in which the transients were below the melting point of the uranium phase. At temperatures above the melting point of the uranium phase, an upward displacement of the fuel core occurred inside the jacket. This movement is believed to have been caused by sodium vapor pressure. Small dimensional changes were noted in the specimens subjected to the more severe transients. The resultant volume changes were less than 1%.

Also on the transients in which the uranium phase was molten, a reaction appears to have occurred between the core and the jacket. Metallographic examinations are under way to verify this conclusion. The results of this series of tests are tabulated in Table XXIX.

Table XXIX. Results of Elevated Temperature Transients on Th-22 w/o U Specimens

Specimen	Jacket	Max. Jacket Temperature, °C	Condition
152I	Nb-1 w/o Zr	940	No jacket failure. No dimensional or volume changes. No core-jacket interaction.
152H	Nb-1 w/o Zr	1000	No jacket failure. No dimensional or volume changes. No core-jacket interaction.
152K	Nb-1 w/o Zr	1435	No jacket failure. Slight diameter and length decrease with resultant volume decrease of 0.4%. Core-jacket interaction noted.
152L	304 SS	745	No jacket failure. No dimensional or volume changes. No core-jacket interaction.
152M	304 SS	923	No jacket failure. No dimensional or volume changes. No core-jacket interaction.
152N	304 SS	1245	No jacket failure. Slight diameter and length increase, with resultant volume increase of 0.7%. Core-jacket interaction noted.

The overall results appear to be encouraging: the dimensional change was far from catastrophic even when the uranium melting temperature was exceeded, and the prospects are good for developing fuel elements with even less interaction between the core and the cladding.

2. Development of Jacket Materials

a. Vanadium and Vanadium Alloys. Jacket materials for the fuel elements of fast reactors must have suitable high-temperature properties, including strength and integrity, good thermal conductivity, and compatibility with the nuclear fuel and with liquid-metal coolants such as sodium. The jacket materials must also be amenable to fabrication into the desired shapes. Of the 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. Since the optimum titanium content in the alloy is not yet known, the vanadium-titanium alloys are 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.

Various molybdenum-base and niobium-base alloys that are commercially available and are considered to be refractory metals are still being investigated.

Consolidation and fabrication of vanadium-20 w/o titanium (TV-20) are continuing. Efforts are being made to encourage industrial participation in the production of alloys of this type. Approximately 34 kg of TV-20 alloy in the form of three ingots, each 10 cm in diameter, has been procured from Union Carbide Corp. (Stellite Division). The ingots were produced by consumable-electrode arc melting of electron-beam-melted ingots, and they are now being evaluated for adherence to specifications.

b. Molybdenum Alloys. The tensile properties of TZM (Mo-0.5 w/o Ti-0.08 w/o Zr) appear to be extremely sensitive to carbon content. Specimen rods machined from wrought vacuum-arc castings, which contained 120 and 170 ppm carbon, and stress relieved at 1200°C for one-half hour, were tested at room temperature and at 650°C. The results are shown in Table XXX. The TZM specimens containing 170 ppm carbon had considerably higher ultimate tensile strengths and yield strengths than those containing 120 ppm carbon. There appeared to be no significant differences in elongations and reduction of areas for the two compositions.

Table XXX. Effect of Carbon on Short-time Tensile Properties of Vacuum Arc Cast Wrought TZM (Mo-0.5 w/o Ti-0.08 w/o Zr) at a Strain Rate of $2.1 \times 10^{-1} \text{ sec}^{-1}$

Temp. (°C)	Ultimate Tensile Stress (kg mm^{-2})	Yield Stress ^(a) (kg mm^{-2})	Elongation ^(b) (%)	Reduction of Area (%)
120 ppm Carbon				
RT	77.6	77.5	(c)	(c)
RT	79.2	63.0	2.0	1.9
RT	79.5	75.5	19.5	18.3 ^(c)
RT	83.7	74.2	12.5	10.5
650	56.0	49.1	25	86
650	57.4	55.0	22	89
650	55.7	55.0	17.7 ^(d)	85
170 ppm Carbon				
29	100	95.2	-	71
650	68.8	66.0	19	84
650	70.1	69.0	21	86
650	72.4	72.4	19	82
650	74.9	72.1	22	87

^aMeasured at 0.002 strain offset.

^bMeasured as change in 2.54-cm gauge length after specimen is removed from furnace.

^cTest terminated before specimen failure.

^dMeasured as change in total length of reduced cross section (4.00 cm).

The effect of strain rate on tensile properties of TZM was studied by pulling some of the specimens that contained 170 ppm carbon at different crosshead speeds. These data are shown in Table XXXI. There does not appear to be an appreciable effect until the crosshead speed reaches a value of 51 cm/min. At this speed, the apparent values of the ultimate tensile strength, yield strength, elongation, and reduction of area all decrease. The calculated strain rate, taken as the ratio of the crosshead speed by the length of the specimen reduced section, corresponding to 51 cm per minute, is $2.1 \times 10^{-1} \text{ sec}^{-1}$. It is doubtful that the Instron tensile machine actually achieved this rate even though the crosshead is servo driven. The mechanical extensometer system was unable to follow the strain at this speed.

Table XXXI. Effect of Strain Rate on Short-time Tensile Properties of Vacuum Arc Cast Wrought TZM (Mo-0.5 w/o Ti-0.08 w/o Zr) at 650°C

Ultimate Tensile Stress (kg mm^{-2})	Yield Stress ^(a) (kg mm^{-2})	Elongation ^(b) (%)	Reduction of Area (%)	Cross Head Speed (cm sec ⁻¹)	Strain Rate (sec ⁻¹)
68.8	66.0	19	84	8.4×10^{-1}	2.1×10^{-1}
70.1	69.0	21	86	8.4×10^{-1}	2.1×10^{-1}
72.4	72.4	19	82	8.4×10^{-1}	2.1×10^{-1}
74.9	72.1	22	87	8.4×10^{-1}	2.1×10^{-1}
76.1	75.5	20	87	8.4×10^{-1}	2.1×10^{-1}
73.4	72.2	20	82	8.4×10^{-1}	2.1×10^{-1}
72.2	70.5	22	87	8.4×10^{-1}	2.1×10^{-1}
55.4	53.4	12.5	89	8.4×10^{-1}	2.1×10^{-1}

^aMeasured at 0.002 strain offset.

^bMeasured as change in 2.54 cm gauge length after specimen is removed from furnace.

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. However, difficulties have been experienced in making stoichiometric monocarbide fuels, and cladding incompatibility is one of the disadvantages often encountered with hyperstoichiometric compositions. 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 investigation and development of methods for production of plutonium monocarbide is essentially complete. The production of solid-solution uranium-plutonium carbide by a diffusion process is being investigated. The powders of uranium carbide and plutonium carbide are being used as a starting material. As previously reported, 20% PuC and 80% UC (-80 mesh) powders, vacuum sintered for 40 minutes at 2000°C, show only small intergranular areas of PuC. It now appears that there is a considerable loss of Pu during high-temperature treatment. One melt charged to produce 19.1 w/o Pu was analyzed to have 17.2 w/o Pu in the 2000°C sintered product. The deficiency is believed to be due to volatilization during vacuum heating. Additional experiments are in progress to determine the extent of these effects and whether they can be controlled by use of a partial pressure of argon gas during the high-temperature heat treatment.

An additional sintering operation was carried out on a PuC-65 w/o UC mixture of -325 mesh powders. The mixed powders were placed in a carbonized tantalum crucible and heated in vacuum to 850°C. Samples were taken after heating intervals of 5, 15, and 30 minutes. Gross weight indicates that some plutonium has volatilized. Analysis is being made for carbon content, plutonium content, and tantalum. Similar experiments will be conducted with helium at one atmosphere pressure during the sintering operation.

4. Development of Powdered Metal Fuel Elements

During this past month, 11 vibratory-compacted, metal-powder-loaded, sodium-bonded, fuel specimens were manufactured. The metal powders were made by micropulverizing U-15 w/o Pu-10 w/o Fz cast material. The uranium was 7.7% U²³⁵ enriched. The powders were classified through 125, 200, and 325 mesh U. S. Standard sieves. Four specimens were compacted from the -120 mesh +200 mesh fraction, four from the -200 mesh +325 mesh fraction, and three from the -325 mesh fraction. The procedure for sodium-bonding consisted of melting a measured amount of sodium over the evacuated and compacted fuel, and pressurizing the system. The amount of sodium added was adjusted to just cover the top of the compact when

theoretically filling the void space between the powders. The final measured sodium level varied from 0.0 to 7.26 mm above the fuel compact. The specimens were capped, closure-welded by the tungsten-arc inert-gas (TIG) method, and cleaned for eddy-current tests. Two of the specimens were sectioned and examined metallographically to evaluate the eddy-current test. Nine of the specimens are now ready for irradiation testing.

5. Development of Fuels for Zero-power Reactors

Very large numbers of small, jacketed, uranium-plutonium alloy fuel plates are required for the study of large plutonium systems in fast critical assemblies. A U-20 w/o Pu-2.5 w/o Mo alloy has been recommended for the additional fuel required for the SEFOR experiments to be performed in ZPR-III. This alloy has good overall properties, and a similar alloy, containing 25 w/o Pu, appears also to satisfy the ZPPR fuel requirements. Metallic fuels are preferred over ceramic fuels because of their higher density, thermal expansion and conductivity. The jackets must be sufficiently durable to remain leak-free over a period of several years of repeated handling, and must fit the fuel tightly so that the linear thermal expansion of the fuel can be transmitted effectively.

A developmental program has been started aimed at developing both a suitable fuel material and the procedures necessary to fabricate and jacket the fuel. It is hoped that these procedures could then be used by a commercial fuel manufacturer to fabricate the necessary critical assembly fuel. The program is divided into two parts. (a) developing a suitable fuel material and a technique for fabricating flat plates therefrom, (b) developing techniques for jacketing the plates.

a. Fuel Development. Five melts were cast during this reporting period. Each was of a U-20 w/o Pu base composition and each contained a different one of the following alloy additions. 1.25, 1.87, and 2.5 w/o molybdenum, 1.33 w/o vanadium, and 1.27 w/o titanium. The alloys were cast in a multiple-cavity graphite mold, internally washed with yttrium oxide. The plate cavities were all 2 in. (5 cm) x 3 in. (7.6 cm) nominal length and width. The plate cavities varied as follows. one cavity 0.086 in. (2 mm) thick, two cavities each 0.177 in. (5 mm) thick, and one cavity 0.250 in. (6 mm). The mold also incorporated two 0.394-in.- (10-mm)-diameter cylindrical cavities.

Specimens have been machined from these melts for further evaluation of metallography, air corrosion, ignition temperature, dilatometry, and compressive strength.

Resistance heating elements are incorporated in the molds, and it was noticed that the end cavities did not fill quite as well as the center cavities. Casting surface finish appears to be a function of the mold-cavity temperature.

The casting dimensions were recorded to compare shrinkage coefficients for the five alloys. Values of from 1.7 to 2.2% shrinkage were obtained for the length, width and thickness. Little difference in casting shrinkage was apparent between the five alloys. Using the average shrinkage value of 1.9% one could predict casting dimensions in the 2.5 w/o molybdenum alloy to within 0.002 in. (0.05 mm) of the length and width, and to within less than 0.001 in. (0.025 mm) of the thickness.

All the specimens were easily machined with unlubricated carbide tools. The titanium and vanadium alloys seemed to corrode more rapidly than the 2.5 w/o Mo alloy, a dark surface appearing shortly after machining. Some difficulty is still being encountered in removing the castings from the molds. It is felt that this difficulty can be overcome by using greater mold drafts or a three piece mold.

b. Jacketing Development. The reference developmental fuel plate design consists of a U-Pu alloy core tightly clad or jacketed in a thin, stainless steel envelope. The half-can jacket was adopted as a design that can be assembled tightly around individual, single-piece, fuel plates and welded without external contamination from the plutonium. The half-cans can be inexpensively cold-formed from 0.3-mm- (0.012-in.)-thick, Type 304 stainless steel.

Two half-cans are pressed over the core in a "bivalve" configuration. A turned-out flange provides metal for a peripheral TIG weld. The seal weld and weld-cast end shoes are formed in the same setup. Filler metal strips are spot-welded to the end flanges. The assembly is clamped between copper chill plates, and the side seal welds are made at one atmosphere helium.

The end shoes are arc-cast with a formed water-cooled chill as a casting mold. Arc-casting is done at $1/3$ atmosphere of helium. Welding at $1/3$ atmosphere prevents a positive gas pressure from developing in the can. The weld castings are shown at the center of Figure 17. The cylindrical castings are then machined flat as shown at the right in Figure 17.

A second shoe configuration has also been tested. This design consisted of two pieces of stainless steel with a machined welding flange. Although this design was acceptable with respect to fabricability, integrity, and service application, it was abandoned because of the high cost of machining the shoe components and the impossibility of decontaminating the fuel plate assembly when alpha contamination appeared in the crevice under the shoe. The applied shoes are shown at the left in Figure 17.

Figure 17. ZPR-III Test Fuel Elements

New dies and weld chills are being machined for production of $5.1 \times 7.6 \times 0.6$ -cm ($2 \times 3 \times \frac{1}{4}$ -in.) and $5.1 \times 15 \times 0.6$ -cm ($2 \times 6 \times \frac{1}{4}$ -in.) stainless steel half-cans. The purpose of these new dies and chills is to further refine the design and increase the core-to-envelope volume ratio. Twenty stainless steel jacketed, natural uranium plates have been made for transient testing in TREAT. The larger U-Pu-Mo cored specimens are being jacketed to determine whether difficulties will be encountered in scale-up.

C. General Fast Reactor Fuel Reprocessing Development

1. Chloride Flux-melt Refining of Uranium-Plutonium-Fissium Fuels

The use of chloride fluxes (halide slagging) for the processing of plutonium-containing fuels is being investigated as an alternative to melt refining in zirconia crucibles. The present studies are being carried out with 80 w/o uranium-10 w/o plutonium-10 w/o fissium alloy, containing about 0.7 w/o cerium as a stand-in for the rare earths. In this process, rare earth elements in the fissium are preferentially oxidized into the flux and thereby separated from uranium and plutonium.

In previous runs, 110 to 200% of the amount of MgCl_2 oxidant required to oxidize the rare earths was generally used. In one previous run, however, no oxidant was added. In this run, only 0.05% of the plutonium was extracted into the flux, while 62% of the cerium was removed (apparently oxidized by impurities in the flux). Although the identity of the oxidizing impurities is unknown, it was considered possible to increase the removal of cerium with an MgO oxidant (or alternatively, to use less than stoichiometric amounts of the MgCl_2 oxidant) and thereby decrease the plutonium extraction (0.3-1.9%) experienced in recent runs with 10 or 100% excess of MgCl_2 oxidant. Accordingly, two runs were made. In one run, the rare earth oxidant was 200% of the amount of MgO required for complete cerium oxidation; in the other run, the oxidant was 80% of the theoretical amount of MgCl_2 . In both runs, flux consisting of 75 m/o BaCl_2 -25 m/o CaCl_2 (mp 875°C) was added in the usual ratio of two parts of flux to seven parts of charge material. Liquation was for one hour at 1150°C. The resulting mixtures were then heated to 1300°C, at which temperature the metal and flux were poured together into a copper mold.

In the run in which 100% excess MgO was added to the flux, a cerium removal of 93% was accompanied by extraction of 0.66% of the plutonium. In the run in which 80% of the theoretical amount of MgCl_2 was added, cerium removal was 93%, and plutonium extraction was 0.10%. These values may be compared with cerium removals of 98-99% in recent runs in which 110 or 200% of the theoretical amount of MgCl_2 was used.

In each run made to date, plutonium vaporization has been less than 0.1%. The results indicate the feasibility of selectively extracting rare earth elements without simultaneously incurring excessive plutonium vaporization or extraction.

2. Skull Reclamation Process

The process for recovery of uranium from skull material remaining in a crucible after melt refining of EBR-II fuel has been demonstrated on a laboratory scale. Engineering-scale (1.5 kg of uranium) runs are now being

made to develop the equipment and operating techniques further. Analytical data have been obtained for waste and product samples from the first two runs in which waste flux was removed just before product dissolution (see Progress Reports for November 1963 and January 1964, ANL-6808, p. 24, and ANL-6840, p. 30). Analysis of the uranium product solutions for the two runs showed that 97.6 and 94.6% of the uranium charged was present and that cerium removals had been very good: 87.5 and 92.3%. The uranium material balances for the runs were 100 and 108%.

In earlier work, flux foamed out of the crucible during skull reclamation runs. Two engineering-scale runs have been performed recently having as one of their purposes the determination of the cause of flux foaming. Observations made during the uranium reduction step of the first run showed that foaming occurred when the melt was vigorously agitated and a flow of argon was maintained through the transfer tube to keep the transfer tube open. In the second run, in which the rate of argon flow during the reduction step was reduced but the agitation rate was unchanged, there was no evidence of flux having foamed out of the crucible. In this run, 88.5% of the waste flux was transferred, one of the highest transfers experienced to date.

3. Materials and Equipment Evaluations

a. High-temperature Materials. Materials are being investigated for the containment of uranium and uranium alloys at temperatures from their melting points to several hundred degrees above their melting points. The test procedure consists of lowering a sample part way into a bath of molten uranium, which is maintained at a desired temperature for the duration of the test. Hafnium carbide, which evidenced little or no attack after exposure to uranium at 1400°C for 24 hr, withstood the test conditions better than the other materials (CrB₂ and TiC) tested.

4. Advanced Processes

a. General. High-temperature processes for purification and recovery of uranium-plutonium fuels from fast power reactors are being developed. Separations are frequently accomplished by using differences in distribution of fuel constituents between liquid metals and molten salts. In this connection, the cadmium-zinc-magnesium system is being studied as a solvent because it may make possible operation at relatively low temperature and the use of stainless steel equipment.

Recent work has been concerned with identifying and characterizing the intermetallic uranium compounds in equilibrium with uranium-saturated, cadmium-zinc-magnesium solutions containing up to 30 a/o zinc and 20 a/o magnesium. It had been shown earlier (Progress Report for April 1964, ANL-6885, p. 17) that the delta phase compound, U₂Zn₁₇, precipitates between about 450 and 600°C and that the substitution of cadmium for

zinc in the U_2Zn_{17} lattice increases with increasing cadmium concentration in the solvent. X-ray analyses of these precipitates show a trend of increasing length of the hexagonal unit cell edges (and volume of the unit cell) as the cadmium and magnesium content of the U_2Zn_{17} lattice increases. The data obtained are consistent with a hypothesis that only two of the zinc atoms in the U_2Zn_{17} lattice can be preferentially replaced. If this is true, the upper limit of substitutional dissolution would be 10.53 a/o. The largest amount of substitutional dissolution (cadmium plus magnesium) observed in the present experiments has been about 8 a/o.

Data have been obtained on the effect of the presence of plutonium on the solubility of uranium in 69 a/o Cd-16 a/o Zn-15 a/o Mg (a solvent composition lying on the ridge of maximum uranium solubility in the quaternary alloy system) at 390 to 690°C. Knowledge of uranium solubility in the presence of plutonium is of practical importance since it is desirable to operate near the maximum fuel solubility, thereby keeping process volumes at a minimum. One run was performed, with plutonium initially present in the ratio $Pu/U = 1/4$. Uranium solubility was found to be essentially unaffected by the presence of dissolved plutonium, although coprecipitation of plutonium with uranium intermetallic compounds occurred to a slight extent.

5. Head-end Treatments for Refractory Fuels

a. Decladding Studies. Studies of the removal of vanadium-20 w/o titanium (TV-20) cladding from fuel by chlorination are continuing. At 600°C, chlorine sparging of TV-20 cladding and uranium-fissium alloy in molten

LiCl-NaCl caused 99% of the cladding and only a few tenths of one percent of the uranium to volatilize. Most of the molybdenum and zirconium (components of fissium) were also distilled. Chlorination of TV-20 in LiCl-NaCl with cupric chloride at 650°C resulted in volatilization of 95% of the TV-20. Chlorination of TV-20 and uranium in LiCl-NaCl-MgCl₂ at 550°C with cupric chloride resulted in volatilization of 95% of the TV-20 with no uranium loss.

D. Sodium Coolant Chemistry

1. Studies of the Oxygen Content in Sodium

Analytical studies of oxygen concentration in sodium are continuing in two sodium-filled loops. The first, a high-temperature test loop (Building 308) operated at 1135°F, is operated primarily for corrosion studies for the Metallurgy Division. This loop has been run for several hundred hours with no change in sodium metal, but has been partially scavenged from time to time by zirconium turnings. Corrosion samples are inserted into the loop through an inert gas lock. Sodium samples, withdrawn at the time of inserting various sets of samples, have been analyzed chemically for oxygen content, which has been found to be run from 100 to 300 parts per million. Recently, a bypass plugging valve loop has been installed on the main loop. While the system was circulating at 660°F, the plugging valve indicated a plugging temperature of 320°F, corresponding to an oxygen concentration of approximately 50 ppm. Sodium flow rates through the valve were varied over a range of two to one ratio with no apparent change in plugging temperature.

At least two factors may be contributing to this discrepancy of data. (a) In this very old sodium, alkaline oxides other than sodium may be appearing in the chemical analysis. (b) In a plugging valve, the crystals must be large enough to wedge in the grooves. For a crystal to grow to such size, the temperature must be below the true point of crystal nucleation.

In the bypass plugging valve loop operating at 400-600°F, both a plugging valve and a distillation apparatus are used on the same loop. The plugging valve has ten grooves, 0.031 in. wide by 0.016 in. deep, cut in the stem cone. The sodium at 400°F temperature and at the start of the run was pumped through the seated valve at calculated linear velocities of 852, 1082, and 1280 ft/min. The plugging temperatures at these three pumping speeds were 255, 251, and 255°F. This second set of data also indicates that liquid sodium velocity through the valve at a given set of conditions does not materially affect the plugging temperature. It was noted, however, that the plugging indication of the x-y plotter was sharper when the linear speed through the valve was higher. This sharper behavior would also indicate that under these conditions of temperature and flow, the plugging is due to crystal size and not to two or three crystals wedging in the grooves. It may be that at slower sodium velocities through the valve, crystals do wedge in the grooves, thus giving a different plugging indication than above.

Distillation runs carried on simultaneously with plugging show scattered results somewhat higher than the 255°F plugging temperature shown above (corresponding to about 35 ppm oxygen). Two sources of error have been found in the distillation method of analysis. Handling procedure has improved but has not entirely corrected the errors. These errors are as follows: (a) Sodium meniscus in the sampling cup which alters the sample volume. (It is hoped that controlled vibration may correct this condition.) (b) Spattering of sample from the cup during distillation. The amount of spattering was indicated by charging the sampling cup with a weighed amount of cupric oxide or silver oxide after withdrawing the sample from the loop. The metallic oxide addition gave an oxygen content of 400-500 ppm. The sodium oxide content was determined by titration in the normal manner. Lower temperature of distillation with attendant longer time has helped the analytical accuracy of the distillation procedure.

The second loop (Building 206) on which analytical studies of oxygen concentration are continuing is operated exclusively for these studies. Initially, this loop was not fitted with a cold trap. The oxide content, as measured, appeared to be unusually excessive (in one case, 860 ppm). A provision was made to scavenge the oxide by the insertion of an improvised cold trap, which consisted of a stainless steel wire basket filled with SS mesh inserted in the flowing sodium stream so that the flow is diverted into a cold leg of the loop. The sodium oxide crystallizes out of the sodium in this leg. A several-day operation of the loop with this insert reduced the oxide concentration level to 55 ppm.

2. Development of Methods of Analysis of Oxygen in Sodium

Conventional methods for the analysis of oxygen in sodium involve the separation of the bulk of sodium from the oxygen-containing compounds and analyzing the residue for oxygen or sodium, assuming that the residue is Na_2O . The difficulties encountered in these methods are those inherent in procedures that require a small amount of one substance to be separated from a large excess of a very reactive constituent. Conventional methods appear to give qualitative results in the range of 10 to 15 ppm oxygen and quantitative results above 50 ppm oxygen.

The development of methods for the analysis of oxygen in sodium has been undertaken at Argonne in an effort to gain greater accuracy and to reduce the limit of detection. Two activation methods are being studied. The first method involves the production of fluorine-18 from the oxygen in the sodium sample by triton irradiation. The tritons are produced in situ by the reaction of thermal neutrons on a small amount of lithium added to the sample, that is, by the ${}^6\text{Li} (n, T) {}^4\text{He}$ reaction. The major problem encountered in this method is that the addition of lithium to the sodium sample causes segregation of the oxygen as lithium oxide.

Because of this difficulty in the triton-activation method, emphasis is being placed on the second activation method under consideration. This method depends on the production of nitrogen-16 by the fast neutron irradiation of oxygen by the reaction $^{16}\text{O} (n, p) ^{16}\text{N}$. The principal problem encountered in the development of this method has been the difficulty of procuring a sample container such that its own oxygen content does not overwhelmingly interfere with the analysis of the oxygen in the sodium. In an effort to circumvent this difficulty, tests are being made to establish the feasibility of irradiating a sodium sample while it is located at one end of a container and counting the sample while it is located at the other, unirradiated end of the container. Blank runs are being performed with a 20-cm-long polyethylene container which will accommodate 8-mm-diameter by 10-mm cylindrical samples of sodium. The results of these tests are not yet available.

E. EBR-II

1. Reactor Plant

The low-power experiments prior to approach to power were begun on May 22. After completion of preparatory work (which included insertion of a second antimony/beryllium source in the inner blanket region), 11 enriched subassemblies were incrementally loaded into the reactor. The current loading consists of 71 enriched subassemblies, including 11 control and two safety rods, and one boron oscillator assembly in place of the 12th control rod. The reactor was made critical on May 25 with approximately 250 lh excess reactivity available.

a. Fuel Handling. The three sensing rods positioned in the primary tank immediately above the three circles of holes in the storage rack are no longer used. Their functions are now accomplished by the vertical movement of the transfer arm and by limit switches actuated by the storage rack drive shaft. This revised arrangement has made possible a substantial improvement of the alignment between the transfer arm and the storage rack at elevated primary tank cover temperatures. This was done by adjusting the rack orientation and transfer arm detent location; however, these realignments resulted in unreliable operation of the sensing rods. Their elimination has allowed the best possible mating between the transfer arm and the storage rack.

To prevent a recurrence of the incident in which a subassembly was dropped into the primary tank, the following steps have been taken:

(1) Interlocks are being designed to assist the transfer-arm operator in determining that a valid wiggle test is being performed and to add an automatic partial check of the test to the existing verification push-button circuit.

(2) Interim operating instructions have been put into effect until modifications are complete. Visual inspection of all transfer-arm positioning pins and the detent pin is required to ensure a reliable wiggle test.

(3) Whenever the transfer-arm mechanism is used, an additional transfer-arm sensing test is performed. It is made by activating the sensing pin when the collar of the subassembly is resting in the carrier block of the transfer arm.

During the reactor loading, the fuel handling mechanism performed satisfactorily.

b. Reactor Oscillator. Connection and disconnection of the reactor oscillating rod to its drive caused considerable delay during reactor loading for approach to critical. The work must be accomplished in a narrow area between two control rod drives and was complicated by a sticking sensing rod. The function of the sensing rod is to verify that the oscillator rod has been separated from its drive before the shield plugs are rotated for fuel handling.

c. Primary-tank Temperatures. The primary-tank, bulk-sodium temperature was maintained at 600°F before the low-power experiments, except for a short period when it was raised to 630°F to check out resistance thermometers and to measure the effective primary tank heat capacity. During the low-power experiments, the bulk-sodium temperature is being maintained at $600 \pm 3^\circ\text{F}$.

The base flow rate for the two primary sodium pumps was changed from 18 to 20% of full flow. During the month, the pumps were operated chiefly for training purposes, and 94 hr of running time was accumulated. Since pump reassembly, both pumps have been operated for 281 hr.

d. Safety Rods. The test of the drop-time characteristics of the safety rods was completed. An effort was made to shorten the time elapsed between actuating the scram button to the start of rod movement, which included the delatch time of the safety-rod drive clutch. Eight runs were made with the voltage drop across the clutch magnet reduced to 18 volts. The elapsed time for this operation was shortened from 0.44 to 0.19-0.26 sec.

e. Fuel-unloading Machine. The new immersion pipe-line heater for the fuel-unloading machine was checked out. After a short period of operation, the heater burned out and was replaced. Steel wool packing, which had been used around the heater, was removed in an effort to extend the heater lifetime. The FUM was used to load a beryllium thimble into the reactor. During the operation, the heater performed satisfactorily.

f. Reactor Building Containment. The inflatable seal on the inner door of the personnel air lock ruptured on May 17. The seal was replaced and a leak test performed on the inner door. The observed leak rate was acceptable.

2. Sodium Boiler Plant

The main secondary sodium EM pump was cut from the piping and cleaned. An inspection of the inside of the pump tube revealed a missing spacer. The spacer's welded flange was still attached near the middle of the duct sheet. Failure occurred between the flange and the web.

Both the top and bottom of the tube were dye-penetrant-tested and radiographed. No additional defects were found. (Visual examination during and following removal of the pump tube from the piping system had disclosed two cracks in the top tube sheet and one crack in the bottom tube sheet.) Figure 19 is a picture of one of the cracks taken before the pump tube was cleaned.

Figure 19. EBR-II Main Secondary Sodium EM Pump - Disassembly for Repair.
Close-up of top side of pump tube showing one of several cracks.

A decision was made to attempt to repair the pump tube in the field. This was done by cutting out the defective areas and welding in replacement discs. The repair welds were dye-penetrant-tested and radiographed.

A cracked weld was found by radiography; it was repaired and successfully radiographed. The pump tube was then leak-tested by helium mass spectrometer, and leaks were repaired satisfactorily.

Figure 20 is a picture of the bottom pump stator before cleaning. Both pump stators were cleaned, and "megger" tests of the electrical circuits were made. The top stator is in excellent condition. However, the power windings in the bottom stator at first showed only 100,000 ohms resistance to ground instead of the 200- to 500-megohm resistance expected. The resistance heaters in the stator were energized and the windings heated to a temperature of 280°F. After two days at this temperature, the resistance to ground rose to over 500 megohms.

Figure 20. EBR-II Main Secondary Sodium EM Pump - Disassembly for Repair.
View of bottom stator in place with thermal radiation shield intact.

The prototype 5,000-gpm pump at Argonne, Illinois, was found to have a leak in the pump tube. Therefore, the possibility of using this pump is being reconsidered.

The steam generator was maintained in wet layup, and the sodium in the storage tank was kept at a temperature of 350°F.

3. Fuel Cycle Facility

a. Preparation for Operations. The air in-leakage rate to the EBR-II Argon Cell, which has been filled with argon since February 27, 1964, has been reduced to 0.007 scfm with the cell maintained at a pressure of minus 2.5 in. of water from a previously reported air in-leakage rate of 0.02 scfm (see Progress Report for March 1964, ANL-6880, p 36). A major leak which has now been corrected was found in the blister on the Argon Cell. The concentrations of impurities in the argon are as follows: nitrogen, 4.5%; oxygen, 40 ppm; and, water, 30 ppm.

To be able to test the effect of a dry argon environment on the operation of equipment items destined for use in the EBR-II Argon Cell, an argon atmosphere enclosure system is being installed. Initially, the system will consist of two gloveboxes and an argon purification system which is similar to the one installed for the EBR-II Argon Cell (see ANL-6766, p. 61). One glovebox and the argon purification system have been installed and operationally tested. Minimum concentrations of impurities in the glovebox argon atmosphere were as follows: nitrogen, 0.1%, oxygen, 4 ppm; and, water, 21 ppm. The installation of the second glovebox is in progress. The argon atmosphere glovebox was used to demonstrate the distillation of magnesium-zinc and the consolidation of the uranium product in the skull reclamation process. The distillation and consolidation steps were conducted in a modified melt-refining furnace. In previous tests (before installation of the furnace in the inert atmosphere glovebox), the distillation of Mg-Zn from uranium resulted in a uranium product which was usually oxidized and difficult to remove from the furnace crucible. The oxidation of the uranium was attributed to air and water vapor which had entered the furnace when the furnace enclosure had been opened for loading or unloading operations. In the run conducted in the inert atmosphere glovebox, essentially all the uranium charged (350 g) to the furnace was recovered from a BeO crucible. About 95% of the product was in the form of a single, clean button which had a metallic luster; the rest of the uranium was in the form of small beads which could be easily removed from the crucible.

The residue (skull) remaining in the crucible after melt refining is to be oxidized in a furnace which was recently installed (together with associated supporting equipment) in the EBR-II Argon Cell (see Progress Report for April 1964, ANL-6885, p. 25). The furnace has been satisfactorily operated in three oxidation runs with nonirradiated skulls from melt-refining runs performed in the EBR-II Argon Cell.

b. Fuel-recovery Operations

(i) Behavior of Iodine in Processing and Off-gas Systems. The first of a series of experiments designed to test the off-gas handling components of argon cell equipment was conducted in the melt-refining furnace. In this run, one Curie of I^{131} (as palladium iodide which decomposes at 350°C) was heated in a stainless steel beaker inside a standard zirconia crucible. No fume trap was used for this run, as its primary purpose was to check the adequacy of the in-cell filter. All the gas from the run, except for a one-liter thief sample, was passed through a caustic scrubber located in the gas line between the subcell vacuum pump and the holding tank. The oil from the vacuum pump was also sampled. The thief sample was drawn from the gas line between the bottom of the feed-through and the vacuum pump. In-cell monitoring, using remotely operated radiation detection instruments, was utilized to indicate the progress of the run and to indicate the disposition of the iodine on the furnace components and on the in-cell filter.

Continuous monitoring of the subcell components with G-M tubes (0.2 mr/hr full scale) failed to show any activity within the sub-cell components. Instruments were placed adjacent to the feed-through, adjacent to the gas discharge line, and at a distance from the feed-through equivalent to a central position among the injection-casting and melt-refining equipment. Table XXXII summarizes the disposition of the iodine found at the completion of the run.

Table XXXII. Distribution of Iodine in Furnace and Off-gas System

<u>System Component</u>	<u>I^{131} (Corrected for Decay) (mC)</u>
Bell Jar	~700
Base Plate	~100
SS Beaker	8
In-cell Filter	4
Thief Sampler	8×10^{-5}
Vacuum Pump Oil*	7×10^{-3}
Caustic Scrubber	8×10^{-4}

* Contained a small amount of water.

(ii) Slag-oxidation Furnace Tests. Three runs were made in the oxidation furnace provided for conversion to oxide of uranium residues in the melt-refining crucible for recovery by the dragout process. These runs were made using slags of previously unirradiated, depleted, uranium

melt-refining runs to check the operation of the equipment and the procedures. The runs, including the oxide dumping step, proceeded without problems. Table XXXIII summarizes the results.

Table XXXIII. Oxidation of Melt-refining Residues

Run No.	Slag Charged (g)	Oxide Recovered (g)	Conversion to U_3O_8 (%)	
			Based on Weight Gain	Based on Oxygen Consumption
1	450	509	75	75
2	310	372	100	96
3	845	962	100	97.5

c. Lanthanum-139 as Burn-up Monitor. Burn-up data on EBR-II fuels will be required by reactor engineers, physicists, and metallurgists responsible for performance and materials evaluation of the reactor. Established analytical methods for determining fuel burn-up are not applicable to the fuel from this reactor. The change in isotopic uranium composition is not sufficiently large, even at 2% burn-up, to permit burn-up determination to the desired accuracy. At lower burn-up levels, this method is completely unsatisfactory. Cs^{137} , often used as a burn-up monitor, cannot be used for EBR-II fuel because at reactor fuel temperatures a portion of the cesium migrates into the bonding sodium in the annulus around the fuel pin and is subsequently lost during decanning operations. Other established methods are equally unsatisfactory for application to EBR-II fuel and the melt-refining process.

The fission product nuclides Tc^{99} and La^{139} were suggested by R. P. Larsen, Chemical Engineering Division, as burn-up monitors for EBR-II fuel since only these nuclides of some 85 fission products appeared to meet the restrictive requirements for this purpose. Tc^{99} has been investigated by R. P. Larsen's group; La^{139} development work was undertaken by the Analytical Group at Idaho.

La^{139} is not a constituent of the fresh uranium/fissium alloy fuel for the EBR-II reactor. It is a stable fission nuclide produced in high yield (top of fission-product yield curve) and, therefore, should be relatively insensitive to yield variations with changes in neutron spectrum. Neither La^{139} nor its precursors are expected to migrate significantly at the reactor temperature of 600°C. Lanthanum is removed during the melt-refining process and does not become a constituent of the recycled fuel.

La^{139} is determined by a mass-spectrometer, isotopic dilution technique following chemical separation from the bulk of the fission products present in the discharged fuel. To determine the precision and accuracy with

which La^{139} can be separated from EBR-II fuel and measured, synthetic uranium/fissium solutions representing 0.1, 0.3, 1.0, and 2.0% burn-up were prepared and analyzed by this method. Five aliquots at each level were analyzed and the precision and accuracy calculated. The overall precision of the method was 2.4% at the 95% confidence level, with no significant variation with burn-up level. The accuracy was equal to the precision of the determination. The precision determined on the synthetic samples was verified by remote separation and determination of lanthanum on several aliquots of MTR-irradiated, 6%-enriched, uranium/fissium alloy fuel.

Application of La^{139} as a burn-up monitor for EBR-II fuel requires good values for fast-fission yield; present literature values have been estimated from parent-yield figures, assuming no independent yield. A direct measure of fast-fission yield has been made on EBR-I, Mark III, fuel using the mass-spectrometric, isotopic dilution method. A tentative La^{139} fast-fission yield for fully enriched EBR-I, Mark III, fuel, based on the Cs^{137} value for burn-up, is given in Table XXXIV. This yield determination must be repeated on EBR-II irradiated U^{235} and U^{238} as soon as it is available.

Table XXXIV. Lanthanum-139 Fission Yield

Material Analyzed	Run No.	La^{139} Found (μg)	Fast-fission Yield (a/o) ^(b)	Thermal-fission Yield (a/o)
EBR-I, Mark III Fuel	1	5.54	5.3 ^(a)	
	2	5.44	5.3	
	3	5.41	5.2	
Capsule 1	1	30.48		6.39 ^(a)
8.44 mg U_3O_8	2	30.68		6.41
Capsule 2	1	33.04		6.47
8.99 mg U_3O_8	2	33.04		6.47

^a Estimated error: $\sigma \approx 1.3\%$ of determined value.

^b Based on number of fissions determined by R. P. Larsen's Chemical Engineering Division using Cs^{137} as indicator of burn-up.

A direct determination of the La^{139} yield from thermal fission of U^{235} was made from 47% enriched U_3O_8 irradiated in CP-5. The number of fissions was determined from changes in isotopic uranium composition during irradiation. The thermal fission yield of La^{139} is given in Table XXXIV.

d. Installation and Test of Fuel Refabrication Equipment. The processing of enriched EBR-II core alloy was started. All major items of equipment are operable, except the fuel-element assembly machine and the

straightness tester. The assembly machine has been moved into the air cell and aligned. It is being assembled and disassembled by remote control. The electrical check-out has begun, and the machine should be in condition to assemble fuel elements on a test basis about June 1. The testing machine and the air-cooled grapples should be received about June 1.

The injection-casting equipment, furnaces, 20-kg balance, charge table, and crucible-unloading station have produced about five enriched-uranium casting batches. These batches were deliberately made small but yielded approximately 350 castings. A new mold pallet was designed and constructed. It is a low-cost, small-mass, disposable item that can be loaded with molds outside the cell.

One of the two pin-processing machines and its data system are operable and are processing castings. This machine is located at the Argon Cell Face 5 where it can be reached with master-slave manipulators for individually loading molds into the machines. A new feeder magazine has been developed for feeding the second machine. The magazine has been tried in the mockup area, where it worked satisfactorily. This new feeder magazine should make the second pin-process machine, which cannot be loaded by master-slave manipulators, operable.

The settlers and welders are in operation. The number of capacitors in the welding power supply was increased from 10 to 12 so a weld could be made with argon shroud gas equivalent to that formerly made by using helium.

Three of the five leak detectors are also operating. A second push-button station has been installed to allow manual flexing of the "Adiprene" O-rings on the leak-detector heads. These O-rings have shown a tendency to set when inoperative for a prolonged period and must be exercised to restore their resiliency. To reduce leakage, the high-pressure Skinner valves have been modified by installing a metal bushing as a retainer above the plastic valve seat.

A pneumatic drive unit has replaced the solenoid driven unit previously used in one bonding machine. The pneumatic striker can bounce the fuel rods more than 5 cm and is completely controllable by adjustment of pneumatic pressure. The modified bonder appears to be able to remove from under the restrainer the bubbles that were not removed by the electro-mechanical striker. Work is under way to reduce the temperature gradient in the bonding magazine and to improve temperature control.

The bond tester is mechanically operable and an evaluation of its operation can be made now that acceptable rods can be produced.

4. Fuel Examination

Several fuel elements that have been in the reactor are being returned for examination by personnel who originally inspected the fuel rods. One control element, L-412, had been returned and was examined during the reporting period. The following inspection steps were conducted:

- (1) Radiography
- (2) Eddy-current test at 20°C
- (3) Eddy-current test at 200°C
- (4) Remelt and quench of the sodium
- (5) Eddy-current test and radiography.

The eddy-current test indicated that the sodium bond in the annulus between the pin and jacket was still acceptable. Several shrinkage voids were observed in the as-received rods tested at 20°C. These voids, however, disappeared with the remelting of the sodium in the 200°C test. The sodium-level measurements showed the level to be higher than originally measured. The average level of the top of the fuel pin after the final testing step was 0.745 in. (1.89 cm), with a range of 0.87 in. (2.21 cm) to 0.61 in. (1.55 cm). Comparison of the recent X-rays of the fuel pins with those originally taken showed an increase in the gap between the restrainer and top of the pin of approximately 0.10 in. (2.5 mm), which is a measure of growth of the pin. It is believed that the high sodium levels were caused by the gas bubbles that appeared beneath the restrainer on all rods inspected, and by small voids in the annulus.

F. FARET

1. General

The final review of Design Package I (Site Preparation) specifications and drawings has been completed, and the drawings and specifications have been signed by the Laboratory and by the AEC personnel.

A revised draft of the FARET Design Criteria received from Bechtel updates the criteria in the areas affected by development of the design since Title I. Laboratory comments are being prepared.

The Laboratory has reviewed and commented on general specifications that will be common to the reactor vessel, sodium pumps, heat exchangers, etc. This material covered broad matters, such as specifications for welding, materials selection, cleaning, and testing.

A draft of the detailed specifications for the reactor vessel is being reviewed. The Laboratory effort on the reactor vessel support cylinder and associated vessel internal components is being accelerated.

The construction contractor, United Engineers and Constructors, Inc., has assigned personnel to work at the Laboratory for closer coordination during the next few months. Emphasis will be placed on the planning of construction activities and coordination of these activities with design and procurement schedules. The work to date has been primarily concerned with schedule analyses, cost estimating, and planning of construction activities.

2. Safety Analysis

The FARET Preliminary Safety Analysis Report (ANL-6813, approval copy) was submitted to the Commission on May 22.

3. Fuel-handling Machine

The essential components of the fuel-handling machine (Figure 21) have been established.

A large pipe serves as the backbone for the traveling bridge, which spans the cell and supports a trolley. Round bars attached to the bridge serve as rails for a trolley on which the machine is carried. This arrangement has the advantage that the area occupied by the structure (in vertical projection) is relatively small so that interference with the TV camera carrier, manipulator, and crane is held to a minimum.

The bridge is propelled by a gear motor with position indication derived from a stationary rack. The trolley can be moved across the traveling bridge to position the handling machine over fuel positions in the reactor grid, the fuel port, the wet fuel storage, or the disassembly machine. These actions could be programmed by automatic controls (situated outside the cell), such as for operation in prearranged sequences, for following established paths between location, or for preventing occupation of certain locations by the machine.

Attached to the trolley is a sleeve in which the handling device is mounted such that it can be moved vertically through a distance of approximately 6 ft. Two pairs of shutters are located at the lower end of the handling device. A hoist drive, sprockets, and a chain retainer are located at the upper end. A chain, weight, and gripper mechanism is used for raising, lowering, and releasing core assemblies. The speed of raising or lowering assemblies is approximately 2 in./sec.

The pulling or lifting force acting on the gripper is regulated and limited by a torque motor.* The downward force is simply and reliably effected by a fixed weight acting on the elevator chain thus preventing excessive downward forces. The sensing and regulating of the performance can be effectively monitored by adjusting the motor current outside the cell.

*A modified, three-phase, squirrel cage, capable of maintaining stalled torque for extended times.

Figure 21. Fuel Handling Machine

The gripper-elevator has been fitted with appropriately shaped parts so that smooth engagement and disengagement are possible, even if the machine and the part to be handled are aligned only close enough to bring their axes within $\frac{1}{2}$ in. of each other.

To handle elements that release appreciable amounts of decay heat, a cooling system will be necessary during fuel transfer. Gas cooling has been selected, at least for the earlier phase of the FARET program, although heat removal is thereby limited to approximately 3 kW. Both once-through and recirculating systems for the gas are being investigated.

4. Component Development

a. Fuel Development. To date, the programmatic development effort for the first zoned core in FARET has involved two likely methods for measuring fuel reactivity coefficients, i.e., the gas-gap and the slip-fit method (ANL-6708). (See Progress Report for February 1964, ANL-6860, p. 38.) The evaluation of experimental requirements and scheduling considerations in conjunction with Laboratory test results has eliminated the gas-gap technique.

The gas-gap method has shown uncertainties of fuel behavior, difficulties with the gas system, complexity of instrumentation and fabricating difficulties due to the vented fuel (see Section b below on "Heat Transfer Bench Tests" and previous Monthly Progress Reports). On the other hand, the slip-fit method offers greater latitude in the range of experimentation and the possibility of obtaining higher fuel temperatures using a mixed oxide fuel.

More specifically, some of the reasons for deciding to abandon the gas gap are:

1. Providing a system for changing the gas pressure and/or composition in the gas gap requires substantial additional development work.
2. Gas lines connected to the fuel elements present problems related to plutonium containment during the manufacture, transportation, installation, and removal of the elements. (Sealing instrumentation in the slip-fit elements presents a lesser problem.)
3. The gas-gap element requires the use of plutonium-uranium carbide, which may be difficult to obtain when needed.
4. The temperature changes observed with the gas-gap element have not yet met expectations. On the other hand, considerable data are available concerning temperatures obtainable with slip-fit elements. (See WAPD-228, August 1960.)

Because of these reasons, it is thought that the slip-fit fuel can be more readily developed on schedule. Therefore, work on the gas-gap method has been terminated, and the work is now directed toward the slip-fit method. Much of the development work, such as the CP-5 loop and the high-temperature instrument work, will be continued, but the effort will be reoriented toward the slip-fit method.

b. Heat Transfer Bench Tests. The thermal bench tests of uranium carbide for the FARET Doppler experiment have been continued and concluded. (See Progress Report for February 1964, ANL-6860, p. 38.) Tests UC No. 1, UC No. 2, and UC No. 3, reported previously, demonstrated that the carbide pellets could be exposed to linear heat fluxes of at least 60 W/cm (equivalent to about 100 W/cm with reactor heating) without cracking. Two subsequent tests, UC No. 4 and UC No. 5, have been run using pellets which were provided with holes to permit the installation of thermocouples in the pellets. In this way, the capability of varying the fuel temperature by changing the pressure and/or composition of gas in the gap between fuel and clad could be verified.

(i) Test UC No. 4. Attempts to pull an effective vacuum of about 10 microns Hg in the gas gap were unsuccessful because of the recurring problem of leakage past the Teflon fittings sealing the heater leads. Accordingly, fuel temperatures were changed by replacing argon with helium in the gas gap. The conditions achieved are listed in Table XXXV.

Table XXXV. Conditions during Test UC No. 4

Gap Gas	Linear Heat Flux (W/cm)	Clad Temperature (°C)	Fuel Temperature (°C)	
			Measured	Calculated*
Argon	45	705	960	1170
Helium	46	700	800	890

*Assuming fuel and clad emissivities of 0.7 and 0.5, respectively.

The fuel-temperature change was only about 155°C, compared with a calculated change of about 275°C. Fuel-temperature changes in FARET for the same heat flux would be much greater, because of the substantially lower clad temperature (about 500°C) and consequently lower thermal conductance of the gas gap. The necessity of maintaining the carbide bench test equipment in an inert atmosphere made it unduly difficult to achieve lower clad temperatures during the tests. When the cladding was examined after the test, the clad inner surface was dark and dull from repeated use at abnormally high temperatures. Therefore, it was likely that the clad emissivity was greater than that used in the calculations; this could have accounted, at least partially, for the difference between calculated

and measured temperatures. Accordingly, it was decided to conduct another test, using a new and shiny clad. In addition, greater clearances were provided between the fuel and the dimples on the clad, to reduce the heat transfer through the dimples.

(ii) Test UC No. 5. The conditions obtained during this test are listed in Table XXXVI.

Table XXXVI. Conditions during Test UC No. 5

Gap Gas	Linear Heat Flux (W/cm)	Clad Temperature (°C)	Fuel Temperature (°C)	
			Measured	Calculated*
Argon	44	590	860	1090
Helium	44	560	680	770

*Assuming fuel and clad emissivities of 0.7 and 0.5, respectively.

Again the fuel temperature change was only about 150°C, compared with a calculated change of about 290°C. However, little importance should be attached to these data because of the final condition of the pellet. When the test element was disassembled, two of the eight pellets used in the test had been reduced almost completely to powder, and these two pellets were located where the temperature measurements were made. All the other pellets were in reasonably good condition, except one which had one of its ends reduced to powder. To date, there is no apparent reason why some of the pellets failed and the others did not.

For future work, it is planned to procure slip-fit fuel elements with which the apparent thermal conductance of the fuel-clad interface will be measured in additional, electrically-heated, bench tests. These data will be compared with those given in WAPD-228 and with those found in the FARET in-pile experiments.

c. Thermal and Hydraulic Analysis Work. The spacing of the test zone and control rod elements for the zoned core was checked to ensure that each element in the subassembly or rod housing will be exposed to the same coolant flow rate.

Initial orificing requirements were established for the test zone, buffer, driver, and plenum-flow-control reflector subassemblies, and for control rods for the zoned core. It is planned to operate with 40 psi in the high-pressure inlet plenum at rated flow. This figure satisfies both the minimum pressure limitation of the FARET pump and the maximum pressure limitation of the initial pump for the sodium-proof test loop (see Progress Report for April 1964, ANL-6885). The plenum-flow-control

reflector subassemblies will throttle the pressure and permit the low-pressure inlet plenum to operate at 16 psi at rated flow. The driver subassemblies will receive coolant from the high-pressure inlet plenum. All other units will receive coolant from the low-pressure inlet plenum. Subassemblies now on order will be tested in a water loop to determine the adequacy of the calculated orifice sizes.

Tentative modifications have been proposed to the core support vessel design (see Figure 22). These modifications provide the following features.

Twenty to 25% of the primary system flow is bypassed around the reactor core to reduce the primary system piping temperature when the core outlet temperature is the design value of 1200°F. The bypass flow leaves the reactor high-pressure inlet plenum through the lower adaptors of the bypass reflector subassemblies, and enters the bottom of the annulus between the core-support cylinder and the reactor vessel. Then the bypass flow rises to near the sodium-free surface in the annulus. At that point, the bypass flow enters downcomer pipes which carry it into a large plenum between the core-support vessel wall and the subassembly shielding extensions. In this way, the bypass flow cools the entire wall of the reactor vessel to nearly the reactor coolant inlet temperature. (The reactor vessel and primary system piping can be operated at a higher temperature by reducing the amount of bypass flow.)

Some mixing of the low-temperature bypass flow and the high-temperature coolant leaving the core should occur in the plenum. Also, the plenum will provide intermixing of core coolant. This should attenuate markedly the rate of coolant temperature change (around 400°F/sec) that may occur at the reactor outlet during scram. This will reduce the effect on the permanent piping system of sudden temperature changes or fluctuating temperatures which may accompany the mixing of fluids at different temperatures. The large plenum will also prevent the high-temperature sodium passing upward between the subassembly shielding extensions from contacting the reactor vessel wall.

The range of temperatures over which Doppler coefficient measurements can be made has been investigated using slip-fit fuel elements of about 0.5 in diameter. It was found that such measurements can be made with effective fuel temperatures ranging from at least 850°F (455°C) to 1775°F (968°C).

The number of fuel-temperature sensors which should be placed in the test zone of the zoned core was estimated. To have a standard deviation of less than 5% assigned to the average fuel temperature, it appears that 15 sensors should be placed at each of 12 positions properly located within the core

Figure 22. Flow Distribution in Proposed FARET Reactor Vessel

d. In-core Instrumentation. A flexible electrical cable has been found that is suitable for the FARET in-core instruments. This cable has, and retains, excellent electrical and mechanical properties when subjected to temperatures of 680°C (1250°F). The cable is commercially available from the Boston Insulated Wire and Cable Company (B.I.W.).

The insulating material on each wire is purified fibrous mica, covered with a layer of purified braided mica. These materials are rated at 815°C (1500°F) continuous use. Tracer material with inorganic pigments is used to color-code the individual wires. This material exhibits excellent color fastness at temperatures up to 540°C (1000°F). Wire materials selected for this particular cable are Chromel, Alumel, and Type AISI 304 stainless steel. The overall cover is braided mica, protected by an optional Inconel braid. The cable can be inserted into a leak-tight stainless steel flexible armor for complete protection against the sodium vapor-argon atmosphere.

A sample of B.I.W. Satellite Style IV, insulated, 28-gauge, solid Alumel wire in the form of a twisted pair has been subjected to temperatures as high as 680°C in air. The insulation resistance was measured at various temperatures and is shown in Figure 23. These values are more than suitable for this application. Gross examination of the twisted pair before and after the heating cycle revealed that the color stripe identifying the wire type had completely disappeared. This loss of color occurs at about 540°C and is not detrimental to the performance of the cable. The cable remained flexible and retained good abrasion resistance.

Figure 23
Insulation Resistance of B.I.W. Satellite
Style IV Wire at Various Temperatures

Similar wire has been subjected to nuclear radiation environments.¹ Fast neutron fluxes of 1×10^{12} nV and gamma dose rates of 5×10^7 R/hr were obtained in the Western New York Nuclear Research Center reactor. The Satellite Style wire was subjected to the above dose rates for approximately 6 hr. This results in a total dose of 6×10^{12} nVt and 3×10^8 R. The temperature of the wire was held at approximately 150°C during irradiation. Postirradiation examination showed that the wire was in excellent condition with no change in brittleness or discoloration. The insulation resistance remained unaffected by the irradiation.

A sample of the 32-conductor prototype cable has been ordered so that further tests can be made. It appears that this electrical cable should satisfy the FARET requirements for high-temperature, nuclear-radiation-resistant cable.

The R and D furnace chamber is being fabricated. The equipment needed for final assembly and initial operation of this furnace has been ordered. It is planned that the furnace will be in operation by the end of June.

The application for a precision sensor is being investigated to measure the temperature of the test samples. A small well-type platinum resistance thermometer should be suitable for this application. An accuracy of about 0.1°C at 650°C appears feasible. This type of sensor will also be evaluated for use as an in-vessel instrument for differential temperature measurements in FARET. Specifications for this sensor are being established.

5. Fuel Assembly Sodium Flow Test Facility

During the month all major elements of the loop have been ordered. The pressure vessel, dump tank, and expansion tank are being fabricated and are due at the Laboratory by July 1, 1964. The interim pump is ready for use, and delivery of the permanent 800-gpm pump is scheduled for November 1964. The deep pit in Building 308 is being prepared for mounting the pressure vessel and the dump tank. Electric power transformers are available and are being located in position.

6. CP-5 In-pile Experiment

Since work on the fuel reactivity coefficient measured in the FARET reactor by using the gas-gap method has been dropped in favor of the slip-fit method, the development work including the CP-5 gas-gap irradiation experiment has been terminated. The experimental loop will be readily adapted to the in-pile experiments for the slip-fit method. The objective

¹Results of the First Five Experiments Conducted at Western New York Nuclear Radiation Center, WANL-TME-109 (only information available).

of the test will be to measure the fuel heat conductance at FARET power operating conditions of 300 W/cm. A description of the slip-fit fuel experiment has been completed. The experimental assembly will be installed in the VT-27 position of the CP-5 reactor. The fuel element capsule consisting of UO_2 or PuO_2 will be enriched to about 3%. The fuel clad will be 304 SS with a 0.527-in ID and a 0.593-in. OD. A thermocouple, consisting of tungsten-3 w/o rhenium versus tungsten-25 w/o rhenium wires, enclosed by a 0.062-in. OD tantalum sheath, will be used for measuring the center temperature of the fuel.

The assembly and wiring of the instrument console have been completed and checked for proper operation. Only minor changes should be necessary when the console is used for the slip-fit experiment.

III. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. Gamma Rays Accompanying Inelastic Neutron Scattering

Extensive studies of inelastic neutron scattering have been in progress for a considerable period of time (see Sect. 3 below). This work is providing uniquely valuable information, but it is limited in sensitivity near the thresholds of the various inelastic neutron reactions. This restriction is particularly limiting in the theoretical interpretation that often places considerable importance on this threshold region. To obtain information in this threshold region, the gamma emission following inelastic neutron scattering is being studied. In the appreciable number of nuclei in which the low-lying level structure is relatively simple, this type of gamma-ray measurement can give very good, relatively inelastic, cross-section values. These can be normalized to the previously measured neutron inelastic scattering cross sections at higher energies to yield good values of the inelastic neutron cross sections from threshold to 1.5 MeV. Thus far, the gamma-ray measurements have been confined to the medium-weight structural materials (niobium, zirconium, iron, and copper) and some heavy-nuclei materials such as rhenium and holmium. Preliminary results have been very promising, and the work is being pursued with diligence.

2. Total Neutron Cross Sections

The measurements of total neutron cross sections employing fast time-of-flight techniques, as described in previous reports, are continuing. In addition, a detailed program of total-neutron-cross-section measurements, using a monoenergetic source, has been undertaken. This latter approach is conventional, but it employs the on-line computer system to automate the experiment. In this manner, both the quality and the quantity of the experimental information are greatly improved. The two methods, time of flight and monoenergetic source, are complementary, since the former provides exceedingly good resolution at energies less than 1.0 MeV, while the latter becomes better at energies greater than 1.0 MeV. Typical of the results obtained thus far are detailed total cross sections for aluminum, uranium (nat), sulfur, iron, cobalt, bismuth, and nickel from 300 keV to 1.5 MeV. The measurements will continue on a periodic basis with the objectives of resolving existing discrepancies in the heavy-element region and of providing more detailed resonance information for the lighter nuclei.

Concurrent with the above experimental work, the Reactor Theory Section has developed a least-squares, resonance-fitting procedure which will make it possible to obtain the maximum physical information from the experimental results.

3. Elastic and Inelastic Neutron Scattering

The comprehensive programmatic study of elastic and inelastic neutron scattering from naturally occurring elements is continuing on a routine basis. Results dealing with scattering from gold, mercury, and thallium and from silver, cadmium, and indium have been prepared for publication. Typical values are given in Figures 24 and 25. In Figure 24 the differential elastic scattering cross sections of silver, cadmium, and indium are given in the form of the Legendre expansion,

$$\frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi} \left[1 + \sum_1^5 \omega_i P_i \right].$$

The solid curves on the figure represent the results of calculations employing a spherical optical model with surface absorption. The agreement between theory and experiment is encouraging and supports the general validity of the optical model employed.²

Figure 24. Differential Elastic Cross Sections of Silver, Cadmium, and Indium

²P. A. Moldauer, Nucl. Phys., 47, No. 1, p. 65 (1963).

Figure 25. Inelastic Excitation Functions of Au

Figure 25 shows the inelastic scattering cross section of gold in considerable detail. Many of the excited states illustrated in this figure had not been known previously. The solid curves of the figure represent the results of Hauser-Feshbach calculations, and the dotted curves include the effects of fluctuations in the resonance widths. Neither calculation is in detailed agreement with the experimental results.

The elements gold, mercury, and thallium are progressively nearer the doubly closed nuclear shell at Pb^{208} . Thus a comparison of the above measurements of scattering from these three targets should show the effect of nuclear shell structure on the scattering potential. An examination of the data indicates that this is true. To obtain good agreement between theory and experiment in this case, the nuclear surface absorption must be reduced systematically as the doubly closed shell is approached. Such behavior has been theoretically postulated previously.³

4. Measurements of $\bar{\nu}$

With the recent modification of the basic Van de Graaff accelerator to deuteron beams (see Sect. 6 below), measurements can be carried out in the neutron energy range 3.5-6.0 MeV. Principal measurements being made are the determination of $\bar{\nu}$ for U^{235} , Th^{232} , and U^{238} relative to that of Cr^{252} throughout the energy interval 3.5-6.0 MeV. The equipment and method are similar to those used previously in this laboratory, but the requisite experimental data acquisition, analysis, interpretation, and conversion to $\bar{\nu}$ is now controlled with the on-line computer. Results thus far have given $\bar{\nu}(\text{U}^{235})/\bar{\nu}(\text{Cr}^{252})$ to a statistical certainty of approximately 0.5%. However, some small, but rather complex, corrections must be made to derive the true ratio from the measured value. These corrections are now in progress and it is expected that the final value of $\bar{\nu}(\text{U}^{235})/\bar{\nu}(\text{Cr}^{252})$ will be known to approximately 0.75% throughout the above energy range. Measurements dealing with thorium and U^{238} are proceeding in a similar manner.

³A. Lane, Phys. Rev. Letters, 2, p. 424 (1959).

5. The $\text{Be}^9(\text{d},\text{n})\text{B}^{10}$ Reaction

Preliminary data have been obtained on the relative yields of the various neutron groups from the $\text{Be}^9(\text{d},\text{n})\text{B}^{10}$ reaction. Neutron reactions leading to the ground state and the first seven excited states of B^{10} have been identified. Work is in progress to obtain the angular distributions and yields of the various groups to learn more of the level structure of B^{10} , and to ascertain the value of this reaction as a neutron source. Parallel with the measurements, a program for the computer analysis of the experimental results has been developed and tested.

6. Instrumentation

The on-line computer is becoming an increasingly productive laboratory tool. Current work has included the automation of nuclear experiments, with concomitant gross improvements in speed and quality. This effort toward increasing control of the experimental and accelerator facilities will continue. In particular, the requisite digital-to-analog systems are being installed to provide better accelerator control; an improved display system, including a storage drum, is under construction; a new input-output interface is being fabricated to provide a large increase in input-output options, and lines have been installed to permit operation of the computer remotely from the ANL Tandem accelerator. The goal of achieving superior efficiency and quality in an experimental nuclear laboratory through the use of an integrated digital system on line has been realized for a period of several thousand operating hours.

Recently the basic Van de Graaff accelerator was modified to utilize deuterons in both pulsed and steady beam operation. This transition makes possible some work with higher-energy neutrons, and has been completed without compromising the intense pulsed capability or the proton acceleration characteristics of the machine. The work described in Sects. 4 and 5 above are possible only because of this modification.

7. High-conversion Critical Experiment

The core now loaded in the High-conversion Critical Experimental Facility has an atom ratio of 1.33 (H U^{238}), and a volume ratio of 0.44 ($\text{H}_2\text{O}/\text{UO}_2$). It is fueled with 3 w/o enriched UO_2 , as 0.94-cm-diameter pellets, loaded into 1.06-cm-diameter aluminum tubing. The fuel rods are spaced in a grid having a 1.17-cm triangular pitch. The current core has a height of 122 cm.

The temperature coefficient measured in the above core was found to be $-0.0097 \pm 0.0006\% (\Delta k/k)/^\circ\text{C}$. This is quite similar to results found with earlier cores having the same fuel, but having a 1.27-cm lattice spacing and a core using 5 w/o enriched BORAX-V fuel. These coefficients ranged from -0.0098 to $-0.010\% (\Delta k/k)/^\circ\text{C}$.

The value of the initial conversion ratio (ICR) measurement made on this core was found to be 0.80 ± 0.01 . A slight correction may be necessary after the effective values of the epithermal capture-to-fission ratio (∞_e) of U^{235} for these cores have been recalculated using the GAM-I code.

Several measurements probably will be dropped from the averages (previously reported for ICR) in view of the existence of a systematic variation in results obtained for Np^{239} yields with materials of various enrichments.

Measurements were made using 3% enriched UO_2 pellet slices and slices of foils made of 3% enriched uranium metal, depleted uranium metal, and highly depleted uranium metal. Some of the samples were dissolved and chemically separated, but many of the Np^{239} yields were measured by coincidence counting of the whole foils. Because of fission product activity, it was necessary to correct for chance coincidences in the 3% enriched foils, which consisted of half the total counts. Further, a systematic variation of measured neptunium yield with foil enrichment was found. Since the foils that were dissolved and separated chemically should be free of corrections for fission product activity, there may be sufficient data to show, with statistical certainty, whether the variation is in the counting or in the neptunium formation.

Plots of cadmium ratios (CR) observed in various cores with different activation materials are shown in Figure 26. The plotted data are sufficiently linear to permit accurate interpolation with confidence equal to that of an experimental measurement. The plot is of $(CR - 1)$ versus the ratio of the number of hydrogen atoms to the number of barns (H/σ_T) of 2200 m/sec cross section for absorption of the homogenized core. No corrections were made for thermal flux hardening, local flux perturbations, or self shielding. The simple relationship is not fully understood although it is apparent that the term $(CR - 1)$, or the ratio of sub-to-epicadmium activation, should be approximately proportional to the ratio of thermal (moderated) neutron density to epithermal density. This latter ratio is determined to a considerable extent by the ratio H/σ_T .

8 Ultra-low-energy Proton-recoil Measurements

The ionization, I , created by a particle traversing matter is related to the energy loss of the particle. The energy loss per ion pair, W , is needed to relate energy to ionization; and experimental data on W are not plentiful for energies in the region of 10 keV or less. It is possible to measure W for protons in hydrogen or methane by observing the ionization pulse-height spectrum from a proportional counter that has been filled with either of these gases and exposed to a $1/E$ neutron flux. At low energies (constant, neutron-proton, scattering cross section), the recoil proton spectrum in response to a $1/E$ neutron spectrum can be easily deduced to

Figure 26. Relationship of CR - 1 Value (cadmium ratio - 1) to H/σ_T Ratio in Various Homogenized H_1 C Cores for Various Materials (where H is the number of hydrogen atoms and σ_T is the total absorption cross section in barns for neutrons having a velocity of 2200 m/sec)

also have a $1/E$ shape. Departure of the measured ionization distribution from this shape provides a direct measurement of W ; if $D(E)$ is the number of recoils per unit energy, and $N(I)$ the number of ionizing events per unit corresponding ionization, then

$$N(I)dI = D(E)dE, \quad (1)$$

so that

$$W = \frac{dE}{dI} = \frac{N(I)}{D(E)}. \quad (2)$$

Equation (2) can be used to determine W since $D(E)$ is known and $N(I)$ is measured. To use Eq. (2), the dependence of I upon E is required. This can be obtained using Eq. (1) in the following way:

$$\int_I^{I_0} N(I)dI = \int_E^{E_0} D(E)dE. \quad (3)$$

The integral on the right-hand side of Eq. (3) is determined analytically, since $D(E) = 1/E$. The upper limits, E_0 and I_0 , are chosen to be in the "asymptotic" region; i.e., W is assumed constant when E is in excess of approximately 10 keV, a result consistent with independent measurements.

Figure 27 shows the variation of W with E for protons in hydrogen. Below 10 keV, a rather complicated behavior is apparent, although W does not depart greatly from its asymptotic value (assumed to be 36 eV/ion pair), at least for energies above about 500 V.

Figure 27
Relationship of Energy Loss, W , per Ion
Pair Formed in Hydrogen with the
Energy of the Impinging Proton Particle

Measurements of W using methane show results similar to that using H_2 , except that methane does not show the pronounced peaking near 900 V.

Measurements in both H_2 and CH_4 are roughly consistent with a model in which W becomes infinite for protons having energy less than about 250 eV, and which assumes its asymptotic value is greater than this energy.

B. Theoretical Reactor Physics

1. Doppler Effect Studies

The influence of the flux-weighting factor on the temperature-dependent, effective cross section has caused some confusion in the current studies on the overlap effect. An attempt has been made to investigate the role this weighting factor plays in the calculations of the overlap effect.

If the narrow resonance approximation is valid, the effective cross section of process x within energy interval ϵ extending from E_1 to E_2 is defined as

$$\tilde{\sigma}_x = \frac{\frac{1}{\epsilon} \int_{E_1}^{E_2} \frac{\sigma_x}{\Sigma_t} dE}{\frac{1}{\epsilon} \int_{E_1}^{E_2} \frac{dE}{\Sigma_t}}, \quad (4)$$

where

$$\Sigma_t = \Sigma_b + \sum_{\ell} \Sigma_{R\ell}.$$

Σ_b and $\Sigma_{R\ell}$ are the background scattering cross section and the total resonance cross section respectively, of the ℓ^{th} isotope.

The numerator in Eq. (4) is proportional to the rate of reaction of the process x , whereas the denominator serves as the weighting factor for the integrated group flux. In earlier work, the quantity $1/\Sigma_t$ in the denominator was taken to be approximately $1/\Sigma_b$. This is not quite true, of course, especially for the case of a large fast reactor where a considerable amount of fertile isotope is present. In fact, the denominator is also temperature-dependent. However, whether or not

$$\frac{1}{\epsilon} \int_{E_1}^{E_2} \frac{dE}{\Sigma_t}$$

is used as the weighting factor instead of $1/\Sigma_b$ makes little difference in the calculated Doppler effect, since the Doppler effect is determined by the change in the reaction rate with temperature, i.e., $\tilde{\sigma} \tilde{\sigma}_x$. When this

temperature-dependent quantity is taken into account, the net effect amounts to the renormalization of the effective cross sections at corresponding temperatures.

To avoid confusion in the studies of interference interactions, the quantity

$$\frac{1}{\epsilon} \int_{E_1}^{E_2} \frac{dE}{\Sigma_t}$$

will be treated in the following way.

Define a temperature correction factor f for the integrated flux as

$$f = 1 - \frac{1}{\epsilon} \int_{E_1}^{E_2} \frac{\sum_{\ell} \Sigma_{R\ell}}{\Sigma_b + \sum_{\ell} \Sigma_{R\ell}} dE. \quad (5)$$

It can be seen quite readily that

$$\frac{1}{\epsilon} \int_{E_1}^{E_2} \frac{dE}{\Sigma_t} = \frac{1}{\Sigma_b} : f. \quad (6)$$

Hence, it is most convenient to rewrite Eq. (4) as

$$\tilde{\sigma}_x = \frac{\tilde{\sigma}_x^{(e)}}{f}, \quad (7)$$

where $\tilde{\sigma}_x^{(e)}$ is the "effective cross section" used in earlier work.

The factor f has been evaluated for some typical systems, and the results are tabulated in Table XXXVII.

2. Approximation of Standard Functions

When $(\arctan x)/x$ is expanded as a Chebyshev series in x^2 , 25 terms are required to obtain 20-decimal accuracy over the range $-1 \leq x \leq 1$. The results of approximating $(\arctan x)/x$ by a Chebyshev series in $t = \beta x^2/[1 + (1 - \beta)x^2]$ are shown in Figure 28, where the logarithms of the coefficients of the 5th, 10th, 15th, and 20th Chebyshev polynomials, (i.e., the errors of truncating the series after 5, 10, 15, or 20 terms) are plotted against β . The great improvement obtainable is obvious. For $\beta = 1.437$ (approximately optimum), 18 terms are needed to attain 20-decimal accuracy, while 24 terms give 28 decimals. A set of coefficients accurate to 27 decimals has been computed on the IBM-1620 computer.

Table XXXVII. Correction Factor f^* A. $\text{Pu}^{239}:\text{U}^{238}$ Systems (s-wave only)

Energy (keV)	$\text{Pu}^{239}:\text{U}^{238} = 1:5$			$\text{Pu}^{239}:\text{U}^{238} = 1:7$			$\text{Pu}^{239}:\text{U}^{238} = 1:10$		
	300°K	750°K	1500°K	300°K	750°K	1500°K	300°K	750°K	1500°K
12.05	-	-	-	0.92655	0.92035	0.91586	-	-	-
6.5	0.90827	0.89899	0.89177	0.91555	0.90640	0.89912	0.91929	0.91005	0.90285
2.25	0.88933	0.87509	0.86284	0.90045	0.88623	0.87441	0.90629	0.89248	0.88038
1.4-2.0	0.87432	0.85736	0.84238	0.88644	0.86995	0.85515	0.89287	0.87655	0.86177
1.0-1.4	0.90254	0.89227	0.88410	0.91665	0.90721	0.89956	0.92420	0.91515	0.90765
0.55-1.0	0.87147	0.85380	0.83743	0.88693	0.87040	0.85471	0.89536	0.87935	0.86387
0.30-0.55	0.89691	0.88460	0.87377	0.91503	0.90492	0.89576	0.92530	0.91620	0.90777
0.10-0.30	0.82780	0.81075	0.79298	0.84659	0.83205	0.81643	0.85759	0.84436	0.82978

B. $\text{U}^{235}:\text{U}^{238}$ Systems (s-wave only)

Energy (keV)	$\text{U}^{235}:\text{U}^{238} = 1:5$			$\text{U}^{235}:\text{U}^{238} = 1:7$			$\text{U}^{235}:\text{U}^{238} = 1:10$		
	300°K	750°K	1500°K	300°K	750°K	1500°K	300°K	750°K	1500°K
12.05	0.92080	0.91467	0.91022	0.92649	0.92033	0.91586	0.92935	0.92318	0.91870
6.5	0.90769	0.89859	0.89148	0.91529	0.90614	0.89899	0.91912	0.90995	0.90277
2.25	0.88695	0.87345	0.86161	0.89931	0.88576	0.87383	0.90563	0.89203	0.88005
1.4-2.0	0.87099	0.85510	0.84070	0.88482	0.86887	0.85434	0.89192	0.87591	0.86130
1.0-1.4	0.89721	0.88846	0.88122	0.91397	0.90537	0.89817	0.92261	0.91405	0.90684
0.55-1.0	0.86289	0.84750	0.83265	0.86246	0.84724	0.83232	0.89264	0.87744	0.86246
0.30-0.55	0.88043	0.87165	0.86378	0.90605	0.89811	0.89063	0.91961	0.91199	0.90464
0.10-0.30	0.79982	0.78637	0.77249	0.82990	0.81802	0.80495	0.84638	0.83518	0.82241

*The U^{238} parameters used in the resolved region were obtained from Firk, F. W. K., Lynn, J. E., and Moxon, M. C., Nuclear Phys., 41, 11.614-629 (1963).

Figure 28

Plot of Logarithms of Coefficients of
5th, 10th, 15th, and 20th Chebyshev
Polynomials Versus β

It is planned to continue these investigations on other common transcendental functions for which the Chebyshev series in the original variable converge slowly.

3. Relative Calculations of Prompt-neutron Lifetimes of Bare and Reflected ZPR-III Core Compositions

Calculated prompt neutron lifetimes of bare core compositions have been compared with analogous calculations of reflected ZPR-III cores. The ratios of reflected-core, to bare-core, prompt neutron lifetimes were calculated for the U^{235} fueled ZPR-III Assemblies 14, 22, 23, and 32. The calculated ratios were 1.04, 1.10, 1.24, and 1.16, respectively. The primary core diluents were carbon, U^{238} , aluminum, and stainless steel, respectively. In all reflected cases, the high-density U^{238} blanket was used.

C. High-temperature Materials Development

1. Ceramic Fuel Development

Among the fertile and fissile materials being studied or developed for reactor use at a maximum temperature in the range 750° to 2000°C are the ceramic compounds of uranium, plutonium, and thorium, and various mixtures or solid solutions of these compounds. Some of the binary compounds have melting temperatures of about 3000°C . Although most of the work on ceramic fuels has been on the oxides and the carbides, the sulfides, phosphides, and such combinations as UP-US, UC-US, PuS-US, and PuP-UP are receiving considerable attention, and the arsenides, selenides, antimonides, and tellurides will eventually be investigated.

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

a. Thermal-property Measurement of UC-20 w/o US. A thermal-conductivity specimen of UC-20 w/o US, a composition near the limit of homogeneity, has been fabricated. The final dimensions were 7.16 cm in length and 2.25 cm in diameter. The initial attempt to make the specimen was by hot-pressing a composition of 69 w/o of hyperstoichiometric UC, 11 w/o UH_3 (to compensate for carbon in excess of the stoichiometric UC composition), and 20 w/o US that had been ball-milled in xylene for 8 hr. The powder was pressed in a graphite mold at 1.70 kg/mm^2 to a maximum of 1600°C for 40 min. A flowing argon atmosphere was used to protect the

specimens in the press from oxidation, since they were not otherwise sealed from the atmosphere. Because of reaction with the mold, this specimen cracked and was unusable. The fragments were crushed and ball milled again to pass a 400-mesh (U.S. Standard) sieve. This material was pre-pressed in a steel die with 1 w/o of binder and then isostatically pressed to 53.50 kg/mm². As pressed, the density was 70% of theoretical. Sintering was done in vacuum at 1760°C for 2 hr. The sintered specimen had a density 91% of theoretical and had no visible defects. In the uranium carbide used, X-ray diffraction gave reflections for two phases: UC with US in solution, and US with UC in solution. The predominant phase, UC solid solution, had a unit cell size of 4.978 Å. Back reflections from the US solid solution were either diffuse or missing.

b. Properties of Thorium and Uranium Phosphides. The fact that thorium phosphide has a melting temperature of about 3000°C arouses interest in the possibility of using actinide phosphides as fuel and/or breeding materials in reactors at very high temperatures. Before the phosphides of uranium and thorium can be evaluated for reactor use, considerable information must be obtained on the characteristics of uranium phosphide, thorium phosphide and thorium-uranium phosphide. A program has therefore been instituted to supplement the extremely scanty data now available. The oxidation behavior, stoichiometry, and fabricating characteristics are among the subjects in need of investigation. Besides giving basic data, the results of this month's research show that thorium and uranium phosphides are not pyrophoric and can be handled in air.

Thermogravimetric analysis experiments were conducted to gain further information on the oxidation of UP. A heating rate of 5°C/min was used. Oxidation, as represented by weight gain, became rapid above 400°C and proceeded at a steady rate to 575°C, at which temperature the rate increased even further. On the basis of weight gain, the major reaction to 650°C was $2UP + 4.5O_2 \rightarrow 2UO_2 + P_2O_5$ (with the P_2O_5 being present as an amorphous phase containing UO_2). Presumably, the change in rate of reaction at 575°C was due to accelerated oxygen diffusion through the amorphous coating. An unknown oxidation reaction took place between 650° and 750°C. Further oxidation of the UO_2 began at 750°C, and it was all converted to U_3O_8 by 800°C. The total weight gain attained by the sample was 30.55%, just short of the theoretical value of 30.73, corresponding to oxidation of the sample to $2UO_{2.67} \cdot P_2O_5$. Reaction of the U_3O_8 with P_2O_5 resulted in the formation of a crystalline compound, without involving any weight change. On cooling, the sample picked up weight as the compound $(UO_2)_2P_2O_7$ formed below 300°C.

UP powder having an average particle size of about 2.5μ was loaded into a graphite mold for hot-pressing under a flowing atmosphere of argon. A temperature of 1720°C was attained and held for 50 min. The fired specimen was fairly dense, but cracked on cooling, because of

sticking to the graphite mold. The specimen was crushed and ball-milled to pass a 400-mesh screen, and 1 w/o stearic acid-acryloid binder (2:1) dissolved in CCl_4 was added to the powder. The damp powder was first pressed in a steel die at 12.65 kg/mm^2 ; the green specimen was then dried and isostatically pressed at 53.50 kg/min^2 . A sound specimen was obtained after slow heating to 1900°C in vacuum, soaking for 2 hr at this temperature, followed by slow cooling. The UP specimen was 6.06 cm long x 2.235 cm diameter and had a density of 93% of the theoretical value.

Chemical analysis of the thorium phosphide powders gave compositions in the range $\text{ThP}_{0.79}$ to $\text{ThP}_{0.91}$. While the compound is most probably hypostoichiometric, more refinement is needed in the analytical technique before the P/Th ratio can be given with sufficient confidence. Thermogravimetric analysis showed that the compound exhibited a weight gain of 27% on heating to 900°C in static air. This appears to indicate that the material had a composition close to $\text{ThP}_{0.95}$. The only crystalline compound observed in the residue was ThO_2 , the P_2O_5 probably occurring in a glassy phase with some thorium oxide.

Thorium phosphide powders are less stable in air than are UP powders, apparently being more reactive with moisture. When fired alone in a crucible at 1800°C in vacuum, thorium phosphide pellets are of a deep blue color throughout. Preliminary melting point determinations in hydrogen indicated a very high melting point for the material, about 3000°C . Solid solution of thorium phosphide with UP is probably extensive, but requires temperatures above 1800°C to produce much interaction.

2. Irradiation of Refractory Ceramic Fuel Materials

Various refractory ceramic compounds of uranium and plutonium are being irradiated to determine their feasibility as high-temperature reactor fuel materials. Clad specimens of the compounds are being irradiated over a range of burnups and temperatures to test their behavior with respect to swelling, fission gas release, and microstructural changes, and their behavior with various cladding materials.

Three instrumented, temperature-controlled capsules containing six clad specimens each are now under irradiation in the MTR. One capsule, ANL-56-7, contains four specimens of vibratory-compacted physically mixed powders of UC-20 w/o PuC and two specimens of vibratory-compacted PuC powder. All six specimens are clad with 0.23-mm-thick Nb-1 w/o Zr and have a fuel diameter of 3.9 mm. These specimens have achieved estimated burnups of from 7.0×10^{20} to 7.8×10^{20} fiss/cc (24,000 to 26,700 MWD/MT-carbide) at cladding surface temperatures between 550 and 610°C . Target burnup for these specimens is 3.67×10^{21} fiss/cc (100,000 MWD/MT-carbide).

Capsule ANL-56-8 contains four specimens of pressed and sintered US pellets and two specimens of vibratory-compacted physically mixed powders of UC-20 w/o PuC. The specimens are clad with 0.31 mm

Nb-1 w/o Zr, and the fuel diameter is 6.4 mm. The sulfide pellets are helium-bonded to the cladding. The specimens have achieved estimated burnups of from 1.1×10^{20} to 1.7×10^{20} fission/cc at cladding temperatures from 490 to 590°C. Design cladding temperatures are 550 to 650°C, and the capsule will be moved to a higher flux position to obtain the desired temperatures. Target burnup for these specimens is 5.50×10^{21} fission/cc (187,000 MWD/MT-US or 165,000 MWD/MT-U for US, and 150,000 MWD/MT-carbide or 120,000 MWD/MT-U for carbide). The difference in MWD/MT values is accounted for by the relative densities of the two types of fuel.

Capsule ANL-56-9 contains an identical specimen loading as capsule ANL-56-8. A W-5 Re/W-26 Re thermocouple was placed inside one US specimen to monitor the central fuel temperature. The capsule was inserted in the reactor this month and has accumulated only negligible burnup. Target burnup for specimens in this capsule is 3.67×10^{21} fission/cc (125,400 MWD/MT-US or 100,000 MWD/MT-carbide) at a cladding surface temperature of 800°C.

3. Anelasticity of Ceramic Compounds

In conjunction with the program for the thorough study of the properties of uranium ceramics, a study has been initiated on the anelasticity of ceramic compounds. Uranium dioxide has been selected as the first base material for the study. Preliminary work has centered on controlling the parameters (including density and grain size) of the UO_2 , and on obtaining the necessary equipment for measuring such properties as elastic modulus and internal friction. These properties are influenced by grain-size, which, in turn, is known to be influenced by such additions as Nb_2O_5 and TiO_2 .

a. Control of Sample Parameters. During the period covered by this report, the effect of minor additions and sintering temperature on the density and microstructure of UO_2 was investigated. Pellets with 0.1, 0.2, 0.3, 0.4, 0.5, 1.0, 1.5, and 2.0 w/o of Nb_2O_5 and 0.1, 0.2, and 0.3 w/o of TiO_2 were prepared by cold pressing with the aid of 2% of binder, and sintered at 1750, 1650, 1550, and 1450°C for 4 hr in hydrogen atmosphere.

(i) Effect of Nb_2O_5 and Sintering Temperature. The results of the density determinations are shown graphically in Figure 29. The density for pellets sintered at 1750°C increased for additions of up to 0.4% of Nb_2O_5 and then decreased; for the specimens sintered at 1650 and 1550°C, the density reached a maximum and then remained constant. The density of the specimens sintered at 1450°C reached a maximum with an addition of 0.3% Nb_2O_5 , decreased slightly with further amounts of Nb_2O_5 , and then remained constant.

Polished sections of the samples sintered at 1750° and 1650°C were made. The variation of grain size expressed in arbitrary units is shown in Table XXXVIII. The addition of Nb_2O_5 up to 0.4 w/o

Figure 29. Effect of Nb₂O₅ and Sintering Temperature on Density of UO₂

Table XXXVIII. Relative Grain Size of UO₂ Bodies with Nb₂O₅ Addition Sintered at Different Temperatures

Specimen	UO ₂ (w/o)	Nb ₂ O ₅ (w/o)	Sintering Temp. (°C)	Grain Size*
1NU	100	-	1750	1
1N1	99.9	0.1	1750	1
1N2	99.8	0.2	1750	2
1N3	99.7	0.3	1750	3.5
1N4	99.6	0.4	1750	3.5
1N5	99.5	0.5	1750	4
1N6	99	1.0	1750	4
1N7	98.5	1.5	1750	4
3NU	100	-	1650	0.7
3N1	99.9	0.1	1650	0.6
3N4	99.6	0.4	1650	1.8
3N5	99.5	0.5	1650	2.0
3N7	98.5	1.5	1650	2.0
3N8	98.0	2.0	1650	2.0

*The grain size of sample 1NU was taken arbitrarily as unity value.

increased the grain size. Larger amounts of Nb_2O_5 did not produce any appreciable change. The samples sintered at 1750°C with 0.6% Nb_2O_5 or more and had a liquid phase present. This phase was not detected in the pellets sintered at 1650°C . The polished sections indicated that the porosity was very low in all the samples. The following mechanism of densification can be suggested by correlating the density and microstructure. The decrease of the amount of grain-boundary area increased the density of the body because the boundaries are less dense than the grain itself. Grain growth probably sweeps out some of the voids that cause the lower density. This suggestion stems from the fact that density and grain growth followed a very similar pattern. This relationship was not observed in some of the specimens sintered at 1750°C , but that can be attributed to the presence of the liquid phase mentioned above.

(ii) Effect of TiO_2 and Sintering Temperature. The variation of density as a function of TiO_2 addition and sintering temperature is shown graphically in Figure 30. The TiO_2 was deleterious for the pellets sintered at 1750° and 1650°C because of a tendency to decrease the density, but when the sintering temperature was lower, the addition of TiO_2 produced denser bodies. They were free of the bloating observed in compositions containing 1, 1.5, and 2 w/o of TiO_2 .

Figure 30. Effect of TiO_2 and Sintering Temperature on Density of UO_2

Polished sections are being made. It will be interesting to see if the specimens that showed a decrease of density with increasing amount of TiO_2 have a liquid phase present.

4. Measurement of Elastic Moduli of High-temperature Materials

Measurements are being made of the temperature dependence of the elastic moduli of materials that show promise in high-temperature reactor applications. To obtain these data, a vacuum furnace has been constructed in which solid bar-shaped samples will be mounted. Pulsed ultrasonic waves will be propagated along the length of these samples, and the transit times of pulses passing a heated, indexed region in each bar will be measured. The indexing is done by machining one end to a smaller diameter than the rest of the bar.

The velocity of sound in the indexed region can then be calculated if the length of this region and the transit time in it are known. Both shear-wave velocities and longitudinal velocities are determined.

Some difficulty has been encountered in obtaining a sufficiently low temperature gradient over the indexed region. Rearrangement of the winding of the heater in the furnace did not reduce this temperature gradient. Since the temperature gradient varies with the temperature of the specimen, a continuously variable adjustment of heat input or heat flow is needed. Attempts to control heat flow out of the furnace by varying the flow of cooling water were unsuccessful. In an attempt to control heat input, a secondary furnace winding will be installed. This winding will be controlled separately from the main furnace winding.

It is often necessary to have a high-voltage signal generator connected directly to a sensitive receiver. This happens when one ultrasonic transducer is used to generate sound in a sample and also to receive echoes from the sample. In these instances, the receiver may be overloaded or may have an overload recovery time that is too long to receive the first few echoes returning through the sample. This problem occurs mostly with wide-band amplifier receivers, rather than in tuned amplifiers. Some time ago, a crystal switch was purchased that would isolate the sensitive receiver from the voltage source during the time interval in which the high voltage was present. To actuate this switch at the correct times and for the proper duration, a driving circuit is needed. A suitable circuit has been developed for use with the Arenberg Pulsed Oscillator and the Sperry Ultrasonic Attenuation Comparator. A circuit for use with the Spike Generator is now being designed.

D Other Reactor Fuels and Materials Development

1 Nondestructive Testing

a Ultrasonic Imaging. Investigations of the angular positioning of relatively thin metallic inspection objects have indicated that the discrete angular positions at which optimum ultrasonic imaging occurs can be correlated with the generation of Lamb waves. Tests with thin plates of aluminum, stainless steel, and brass in various thicknesses, have shown excellent correlation with calculated angles for the generation of Lamb waves. Further tests with such samples have shown that in all cases, first-order modes are preferred for imaging, probably because of increased intensity. In some cases, such as those in which the asymmetrical mode velocity approaches that of surface waves, a clear choice between first-order modes for optimum imaging can be made. In the case of the example cited, the symmetrical mode yields a much improved result. A report describing these results in greater detail is now in preparation.

Comparison tests of flat fuel elements by ultrasonic and radiographic imaging have been completed. The comparison ultrasonic tests tend to confirm the imaging system results. Several of these plates are now being sectioned for metallographic tests.

The imaging system was used during this period for the through-transmission ultrasonic inspection of several V-20 Ti flat plates. One defect indication was noted in the 12 plates tested.

b Correlation of Heat Transfer and Bond Quality. Preliminary ultrasonic tests on copper braze-bonded specimens indicated the necessity of several changes in the existing testing setup. These samples are not only thin (0.127 cm) but are also only 1.9 cm square. In such a case, the edge region causing sound scattering is a fairly large proportion of the sample size. The Lucite specimen holders were abandoned since they only intensified the scattering effect. The scattering appears to be a minimum from the specimen-water boundary (the specimen is held in a holder only along one edge). Such a setup, however, renders the conventional recording systems useless since it is impossible to gate out the water pulse. Other methods of reading the information are being considered.

c Infrared Imaging. An infrared-sensitive Vidicon camera tube has been placed in operation. Initial efforts with the present system indicate a threshold response of approximately 250°C. This threshold object temperature probably can be lowered by using improved optics and better video circuitry. Efforts to improve these are now under way.

Installation of a 24,000-rpm motor in the mechanical infrared imaging system increased the chopping frequency eight times, and thereby improved the temperature resolution. This was determined by observing

pictures taken with infrared radiation of a hand at body temperature. Previous hand pictures showed only an outline with no evidence of temperature differences. Pictures taken with the higher chopping frequency showed definite temperature differences on the hand. This result indicates a considerable improvement in signal-to-noise ratio. The indium antimonide detector used for the hand pictures has a sensitive area of 2.4×1.35 mm, which gives the system poor angular resolution. The angular resolution can be improved by using a field stop with considerable loss in temperature resolution and low-temperature response.

A lead sulphide detector, with a sensitive area of $\frac{1}{2} \times \frac{1}{2}$ mm, was installed in the imaging system, and a transistorized Z-axis amplifier, with a good low-frequency response, was built for the detector. The detector is used without chopping the infrared signal by scanning the field at a rate of 180 lines per minute. This detector gives excellent angular resolution and has a better low-temperature response than the indium antimonide used with the field stop. A temperature of less than 50°C can be detected with this detector.

E. Remote Control Engineering Development

1. Viewing Systems

Studies have continued on the effect of additives in small concentrations to experimental glasses as part of the program to develop better radiation-resistant glasses. The nature of the ultraviolet absorption produced by small amounts of iron and lead has been determined in an aluminoborate glass melted under normal and reducing conditions. This will be useful in identifying these impurities in high-purity experimental glasses.

Electron irradiations at 10 MeV of small samples of shielding window glasses has been continued as an accelerated test in determining the susceptibility to discharge of experimental glasses when exposed to gamma radiation. Preliminary tests indicate that the rate of dissipation of charge after irradiation is a sensitive parameter.

2. Electric Master-Slave Manipulator Mark E4

Design studies have been continued on the latest configuration of the Electric Master-Slave Manipulator Mark E4. Several different gearing, mechanical cabling, and counterweighting arrangements have been studied. Considerable progress has been made in solving some of the problems; however, a number of knotty problems still remain.

In the new configuration, the servo arms have a mechanical link about 40 in. long between their shoulder joints and the servo drive units. This should make the arms better for reaching in and around machinery.

To improve reach further, studies are also being carried out on providing a small forearm next to the wrist joint. Full-scale mockups have been made of some of the configurations of the master and slave arms to determine the overall configuration most suitable for wall mounting, overhead mounting, or mounting on a wheeled vehicle to form a slave-robot. Only small modifications to the manipulator would be required for each type of mount.

The servo amplifier was previously mocked up and tested. During this month, the components were assembled on a special chassis to study the configuration of the amplifier in preparation to building these in their final form. The component arrangement and the installation of the amplifier in a rack was designed so that it can be either serviced in place or easily removed for replacement. A number of circuits are on plug-in module cards.

3. Special Motors for Master-Slave Manipulators

Work has progressed on the low-inertia motors for future electric master-slave manipulators. The two-phase, 60-cycle motor with a cup rotor is nearly ready for test. All parts have been fabricated, the laminations have been assembled, and the stator has been wound ready for impregnation.

F. Heat Engineering

1. Studies of Boiling Liquid Metals

a Void Fraction - Pressure-drop Facility. This facility is designed to investigate the two-phase flow characteristics of boiling sodium; in particular, experimental information is desired pertinent to the vapor volume fraction and two-phase frictional losses in an adiabatic test section. The loop is constructed of Type 316 stainless steel and is limited to short-term operation at approximately atmospheric pressure and about 1630°F. Considerable effort has been expended in the development of reliable, accurate instrumentation for the measurement of pressure differentials and vapor volume fraction.

The five 10-kc, eddy-current liquid level probes have been calibrated and installed in the loop. The sodium-fill procedure has been completed, and medium-temperature (up to 1200°F) operation has begun. Heat loss, gamma attenuation, and electromagnetic flowmeter calibrations will be undertaken in the next month, before high-temperature boiling measurements

2. Boiling Sodium Heat Transfer Facility

a. Niobium-Zirconium Loop Construction. Detailed drawings of the vacuum chamber lifting fixture, cooling water supply and drain manifolds, manifold stand, and liquid nitrogen manifold were completed and forwarded to the shops for fabrication. An attachment for the lifting fixture for handling the vacuum-chamber oven has been designed, and a preliminary drawing detailing the necessary modifications to the building water and drain piping and high-pressure air supply was completed. The bulk of the remaining materials for the cooling circuits and handling equipment have been ordered, and a small-parts ordering and inventory scheme has been initiated to locate and maintain control over the many items involved.

An elevated walkway to be used in conjunction with vacuum-chamber handling and sodium-loop assembly has been designed and is being detailed. Initial planning of electrical services for the vacuum chamber has been completed; sketches of a recommended conduit layout have been prepared and will soon be submitted for final design of the installation.

The vendor has experienced considerable difficulty in attaching tightly spaced coolant tubes to the upper vacuum-chamber wall. A revised cooling-channel design has been proposed and is being reviewed. Delivery of the equipment has been delayed approximately one month because of this problem.

b. Heater Experiments. Assembly of the small-scale, thermal-radiation-heated experiment has been completed. Leak checking showed the system to be sufficiently leak-tight. The electromagnetic flowmeter has been calibrated and installed. After resolving problems concerning the operation of one of the temperature recorders and after final checkout of the instrumentation, the NaK bubbler in the argon purification system and the supply tank will be charged with NaK.

Installation of the 200-kW power supply to be used in the electron-bombardment heating experiments has been completed. Fabrication of the electron-bombardment-heated sodium boiler is underway.

3. Double-pipe Liquid Metal Heat Exchanger

The design work on the mercury heat-transfer loop described in the preceding report (see Progress Report for April 1964, ANL-6885) has been completed, and the material required for construction of the loop has been ordered.

Because of delivery and fabrication considerations, a replaceable dummy test section of stainless steel will be constructed for the initial

runs. While the actual nickel test sections are being fabricated, the loop will be checked out and calibrated with the dummy test sections.

As described in the Progress Report for April 1963 (ANL-6717, p. 49), an analytical solution has been obtained for a double pipe configuration consisting of a circular tube surrounded by a narrow annular space. For a wide annular-space configuration, a solution involving the annulus ratio as a parameter is required. A plug-flow solution for the wide annulus case has been formulated. A complete investigation of this solution remains to be initiated. It is expected that results similar to those shown in Figure 9 of the Progress Report for October 1962 (ANL-6635, p. 42), and Figure 16 of the Progress Report for April 1963, will be obtained.

As reported in the April 1963 Progress Report, an approximation technique exists to solve slug-flow cases for turbulent flow with Peclet numbers of up to 1000 for the parallel plane and narrow annular-space configuration. This involves a new parameter, k^+ , a dimensionless factor which expresses the effect of turbulence on the effective total conductivity of the fluid. For the parallel plane and narrow annular space configurations, k^+ is a function of the Peclet number (see ANL-6801, p. 55). With the wide annular space, it is expected that the k^+ parameter should be a function of both the Peclet number and the annulus ratio. Relations for k^+ for this configuration are being obtained.

4 ANL-AMU Program

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

a. Propagation of Void Waves in an Air-Water System. The electrical resistivity probe, to be used for measuring voids, produced results that are about 25% lower as compared to other reliable techniques. To use this probe for further studies, it must be calibrated. Since there is no known method for determining accurately local values of void fraction, it must be assumed that the error from the probe is independent of radial position in the channel. This assumption facilitates the calibration of the probe technique.

Correction of the probe error was accomplished by incorporating a variable delay in the "void probe trigger." This addition to the circuit delays the "shut off" of the modified bubble pulse. In this manner, the correction is distributed equally to each bubble. Calibration of the variable delay must be done separately for each channel of the "void probe trigger" because of slight differences in electrical components. Five of the six channels have been calibrated.

Two runs with a square-wave air input were made, and the voids were observed at five different vertical positions in the test section. The signals from the five probes were recorded on magnetic tape. The signals were then sampled by an analog-to-digital converter. The playback speed of the probe signals was varied to note the effect on the results. The main purposes of these two runs are to establish the reliability of the experimental procedure and to check the computer program. No results have as yet been obtained.

b. A Study of Two-phase Flow Parameters during the Complete Vaporization of a Fluid. The construction of the two condensers and the subcooler have been completed. Assembly of the test loop has been delayed because of difficulties encountered with the hangers for the loop. The hangers are to be shipped in two weeks, and the welding of the test loop will be accomplished on their arrival. A testing of the visual test section has been delayed by an absence of thermocouple fittings. Two types of thermocouple assemblies will be used on the external side of the heated test section. In one type, the thermocouple is to be mounted through a single opening and will lie parallel to the test section for $\frac{1}{2}$ in. In the other, one thermocouple lead will enter one hole and extend along the test section and then exit from a hole approximately 3 in. down the test section. Any influence of the electrical circuit should be indicated by this arrangement. Both types of thermocouple settings will be insulated from the heated strip by a thin sheet of mica. Twelve equally spaced holes have been drilled in the test section shell. Thus, 23 thermocouples will be mounted in the unit. Some heating elements in the preheaters have been replaced and the units assembled.

The correlation of surface tension for water and the freons has been extended over a range of reduced pressure (saturation pressure/critical pressure) from 0.001 to 1. The form of the single curve is complex, and it appears that the most suitable representation is graphical. Deviations from the single-curve result do not exceed 10%, and this is within the accuracy of the original measurements.

c. Transient Behavior of Heat Transfer Loop. To evaluate the pressure pulsations impressed upon a venturi flowmeter by a progressive acoustic wave passing through the venturi, the following analysis is presented. The pressure distribution is given by

$$p = P \sin 2\pi (ft - x/\lambda),$$

where

P = amplitude of pressure oscillations (psi);

f = frequency (cps);

t = time (second);

x = axial distance (ft);

λ = wave length (ft).

The pressure difference between the venturi pressure taps whose axial positions are x_1 and x_2 is

$$\begin{aligned} p(x_2, t) - p(x_1, t) &= P \left[\sin 2\pi \left(ft - \frac{x_2}{\lambda} \right) - \sin 2\pi \left(ft - \frac{x_1}{\lambda} \right) \right] \\ &= 2P \sin \left[\pi \frac{x_1 - x_2}{\lambda} \right] \cos \left[\pi \left(2 ft - \frac{x_1 + x_2}{\lambda} \right) \right]. \end{aligned}$$

The amplitude (a) of the venturi pressure oscillations is

$$a = 2P \sin \left[\pi \left(\frac{x_1 - x_2}{\lambda} \right) \right] \sim 2\pi P \left(\frac{x_1 - x_2}{\lambda} \right),$$

and the frequency is the same as that of the absolute pressure.

With the following values which pertain to the Freon-114 loop,

$$P = 20 \text{ psi},$$

$$x_1 - x_2 = 0.208 \text{ ft},$$

and

$$\lambda = 49.1 \text{ ft (see April Progress Report),}$$

the amplitude is found to be

$$a = 2\pi (20) \frac{(0.208)}{49.1} = 0.53 \text{ psi.}$$

This value is in fair agreement with experimentally determined values. However, the experimental values show more scatter than is desirable when the two amplitudes are cross plotted. During the experimental runs, it was noted that any vibration of the venturi pressure lines affected the readings and it was impossible to completely isolate the lines from the vibrating loop. Consequently, the scatter is a reflection of the quality of the measured pressure differentials.

d. Inception of Instability in a Natural-circulation Loop. The construction of the loop and the installation of the necessary instruments are complete. The system is now being debugged.

The details of the instrumentation for this study are as follows: In the present arrangement, there are four chromel-alumel thermocouples located at the test-section inlet, steam separator, condenser outlet, and downcomer. The thermocouples are connected to a high-speed recorder. The power to the test section and the pressure drop across the venturi in the downcomer will also be recorded. Provision is made for two more transducers for measuring pressure drops across the test section and riser, and also for an additional thermocouple.

To prevent burnout, six thermocouples are spot-welded at various positions near the upper bus bar, and the opposing signals are sent to a normally open relay.

G. Engineering Mechanics

1. Switch Contact Resistance in Strain Gages

During an experimental stress analysis of the CP-5 mockup plenum chamber, it became evident that the switching mechanism used with strain gages did not have constant characteristics. Certain contemplated applications of strain gages require extreme accuracy, which make positive operation of the switching mechanism a necessity. A program was initiated to test the accuracy of the switches:

A test of commercial, standard, portable, strain-gage indicator switches was made to ascertain repeatability. Twenty switches (relatively new) were cycled 17 times each over five days in a manner representing standard switching procedure. Switching was also done at random. The switch contact resistance was measured, recorded, and the degree of non-repeatability was calculated and designated as error. The results, based on ohms resistance to four places (0.0000), are as follows:

Lowest mechanical switch error (average) = 2.27%;

Highest mechanical switch error (average) = 5.65%.

The mercury type switch is believed to possess better contact resistance characteristics than the mechanical type switch. Seven different types of magnetic mercury switches were selected for testing at no-current load and 10,000 cycles, from which one type was to be selected for use in a completed switch and used under simulated operating conditions. The results of the product test, based on ohms resistance to four places, are as follows:

Lowest mercury switch error (average) = 0%;

Highest mercury switch error (average) = 5.13%.

The resistance of strain gages is from 60 to 1500 ohms, with the predominance at 120 ohms. The gage factor ranges from 1.5 to 3.5 with the predominance at 2.0 and 2.1.

If the stress in aluminum is measured with strain gages with a resistance of 120 ohms and gage factor of 2.0, and using a mechanical switch with the lowest (2.27%) average error, an error of 121 psi in the measured stress occurs. The 121-psi error for this switch corresponds to an error of 1% when measuring near the ultimate tensile stress of the aluminum and greater than 2.6% for stress below the yield point. The stress error can be reduced by increasing the gage factor and resistance, or by decreasing the error attributed to the contact resistance in switching.

H. Chemical Separations

1. Fluidization and Volatility Separation Processes

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

Laboratory work in support of the pilot-plant study of a fluid-bed fluoride volatility process is directed toward determining the optimum conditions for the fluorination of U_3O_8 - PuO_2 mixtures in a fluidized bed of alumina granules and for efficient removal of plutonium from the fluid bed. The magnitude of the plutonium loss to be expected upon discarding alumina to waste has not been established since the value will depend upon several factors, such as the number of times that the alumina bed can be reused.

The effect of the reuse of the alumina bed material on plutonium retention on the alumina has been studied in a recent fluid-bed experiment. Four additions of PuO_2 -fission product mixture containing 1.22 grams of plutonium and 2.58 grams of selected fission products were made to a single alumina bed. The experiment was performed using a feeding-fluorination period at 500°C with the gas phase containing 20 v/o fluorine in nitrogen, and a recycle-fluorination period of 10 hours at 550°C using 100% fluorine. After fluorination of the first addition, the alumina bed contained 0.0045 w/o plutonium; after the second addition, the bed contained 0.014 w/o plutonium. This level of plutonium retention on the alumina was maintained for the third and fourth additions. At the conclusion of the experiment, 97% of the plutonium added had been volatilized. These data suggested that the feeding of seven batches of U_3O_8 - PuO_2 -fission product mixture to a single alumina bed would result in the retention of one percent or less of the total plutonium. An experiment based on this concept has been carried out. Analyses of plutonium retained in the alumina bed indicate that after seven additions of a U_3O_8 - PuO_2 -fission product mixture to an alumina bed, where each addition was followed by a

period of recycle fluorination, significantly more than 99% of the plutonium charged had volatilized. It is believed that some of the earlier results for this type of experiment, suggesting difficult plutonium removal, were artificial. These results were probably caused by the difficulty in operating a small-scale fluidized bed. This possibility is currently under investigation.

No direct determination of the structure of the plutonium hexafluoride molecule has been reported. However, the similarity in properties between UF_6 , NpF_6 , and PuF_6 indicates similar structures. The uranium hexafluoride structure was shown to be orthorhombic with the D_{2h}^{16} -Pnma space group.⁴ On the basis that the structures of UF_6 , NpF_6 , and PuF_6 are isotypic, the lattice parameters and volumes of the unit cell have been calculated for these compounds from X-ray data of Florin, Tannenbaum, and Lemons.⁵ The lattice constants for PuF_6 are $a_0 = 9.912 \pm 0.004$, $b_0 = 8.942 \pm 0.005$, and $c_0 = 5.206 \pm 0.006$, which result in a unit cell volume of $461.4 \text{ \AA}^3$. Comparison of the unit cell volumes of UF_6 ($466.0 \text{ \AA}^3$), NpF_6 ($463.8 \text{ \AA}^3$), and PuF_6 ($461.4 \text{ \AA}^3$) illustrates the actinide contraction.

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

(i) High-activity-level Facility. A bench-scale facility has been constructed for the study of the fluid-bed chlorination-fluorination process using highly irradiated uranium-Zircaloy or uranium-aluminum alloy fuel materials. The facility, shown in Figure 31, has been designed to operate with up to 10,000 Curies of activity and will be installed in the Senior Cave of the Chemical Engineering Division, Argonne, after brief testing with inactive fuel materials. The tests involving processing of irradiated fuel will define clearly the disposition of fission products and the total decontamination achieved in the process.

The facility consists of a $1\frac{1}{2}$ -in -diameter fluid-bed reactor and a packed-bed filter with associated equipment for the containment of volatile chlorides and uranium hexafluoride produced in the process. Volatile zirconium tetrachloride or aluminum trichloride produced by reaction of the fuel alloy with hydrogen chloride is condensed to a solid chloride which is readily disposed by dissolution in dilute hydrochloric acid and subsequent solidification in cement. The uranium hexafluoride produced by reaction of uranium trichloride with fluorine is decontaminated by a sorption-desorption operation in a series of four sodium fluoride traps maintained at temperatures between 20 and 400°C . The hexafluoride product is subsequently collected in a series of refrigerated traps. Disposal units for hydrogen chloride and fluorine gases are included in the facility. All liquid wastes generated in the process will be solidified with Portland cement before discharge from the hot cell

⁴J. L. Hoard and J. D. Stroupe, Cornell University Reports A-1242 and A-1296, 1944.

⁵A. F. Florin, I. R. Tannenbaum, and J. F. Lemons, J. Inorg Nucl Chem, 2, 368 (1956).

Figure 31

Fluid-bed Volatility Facility for Studies with Irradiated Alloy Fuels (Front view of equipment, before installation in CEN Senior Cave)

All equipment components have been designed for completely remote operation and maintenance. Initial shakedown tests, currently under way, involve evaluating the remote operation of the equipment and performing a cold run with unirradiated alloy containing 3.9 w/o uranium in Zircaloy. After successful completion of the preliminary tests, the facility will be permanently installed in the cave for demonstration of the reprocessing scheme with irradiated fuel.

2. General Chemistry and Chemical Engineering

a. Preparation of Uranium Monocarbide. A batch of uranium carbide was made by adding activated charcoal to uranium dissolved in zinc-14.2 w/o magnesium and stirring at 800°C for 12 hr. A charge of 1000 g of uranium and a C/U ratio of 1.02 were used for the most recent preparation. The oxygen content of the uranium monocarbide product was 0.18 w/o, which is below the allowable limit of 0.2 w/o, however, the total carbon content, 5.24 w/o, exceeded slightly the allowable upper limit of 5.2 w/o.

The first of two runs to produce uranium monocarbide from enriched uranium (93% U^{235}) and activated charcoal was completed. A yield of 388 g of UC from a charge which contained 470 g of enriched uranium was obtained.

3. Chemical-Metallurgical Process Studies

a. Neodymium-Cadmium Galvanic Cell Studies. The thermodynamic functions of the neodymium-cadmium system are being evaluated by means of a galvanic cell. A cell of the type

has been assembled, and the emf has been measured as a function of temperature. Preliminary results indicate that the free energy of formation of $NdCd_{11}$ is about -36.5 kcal/mole at 400°C.

I. Plutonium Recycle Program

1. Plutonium Recycle Experiment

Preliminary calculations were made of the effect of voids, and of a uniform increase in moderator temperature on the reactivity of the initial plutonium critical assembly. In the former case, the voids were assumed only for the plutonium zone, and the system was assumed to be at room temperature. Apparently the coefficient of reactivity due only to a temperature change is much smaller than the void coefficient of reactivity, and the effort involved in experimentally measuring the void coefficient only cannot be justified. The combined temperature-void effect is easily measured using the startup heaters.

The small temperature coefficient of reactivity is due to the small difference resulting from the positive reactivity effect from the thermal group owing to the Pu^{239} resonance at 0.3 Volt, and the negative epithermal effect owing to Doppler broadening of the U^{238} resonances.

The accuracy of the calculational model is being investigated by applying the model to the computation of the criticality of the mixed oxide critical experiments being performed at the Hanford Laboratories. The Hanford criticals have uniform hexagonal lattices with pitches from 0.90 to 0.55 in., a range which includes lattices similar to that of the EBWR recycle experiment. Also, the fuel used at Hanford has the same composition as will be used here at Argonne.

2. Plutonium Recycle Fuel

Fabrication and procurement of the fuel loading for the Plutonium Recycle Experiment are a joint Argonne-Hanford responsibility. The fuel

loading is a three-zone loading with plutonium in the central zone, partly-enriched uranium in the intermediate shim zone, and natural uranium in the outer blanket zone. All fuel elements are designed to contain 36 fuel rods in a square lattice. Each fuel rod contains oxide fuel in 0.945-cm ID x 0.064-cm wall Zircaloy-2 tubing. The loading and composition of fuel for each zone is as follows:

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

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

(c) The blanket zone of 58 elements is fueled with natural UO_2 . This fuel is also being fabricated by United Nuclear Corporation.

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

(a) The plutonium is converted to PuO_2 .

(b) The PuO_2 is blended with UO_2 and compacted using the Dynapak equipment of high-energy-rate-forming, "Nupac," process.

(c) The Nupac fuel and load are crushed and screened, using vertical excitation of the tube, into fuel rods by the vibratory compaction, "Vipac," process.

(d) The loaded rods are sealed, inspected, cleaned, and tested by conventional procedures.

Some of the original plutonium supplied for the plutonium recycle loading was found to be contaminated with halides, which had to be removed. Except for some additional plutonium required for the replacement of contaminated material, all plutonium has been converted to the oxide.

A fused UO_2 was originally used for blending with the PuO_2 . More recently a higher-density high-fired UO_2 has become available and will be used on all subsequent loadings. Approximately one-half of all PuO_2 - UO_2 oxide fuel required for the plutonium recycle loading has been processed through the Nupac step.

A total of 400 central-type plutonium recycle fuel rods acceptable for reactor service have been loaded. An additional 450 central-type rods not acceptable for reactor service have been loaded for use in approach to critical and other physics tests at Hanford. Since 1500 rods are required for some testing, acceptable rods will also be used before delivery to ANL for final fuel element assembly.

Procurement of all necessary hardware for loading all $\text{PuO}_2\text{-UO}_2$ fuel rods has been completed at Hanford.

Most of the components and hardware needed by United Nuclear Corporation for both the enriched and natural fuel element loadings have been procured. Evaluation lots of natural UO_2 pellets, enriched UO_2 pellets, Zircaloy-2 end-plug stock, and Zircaloy-2 jacket tubing have been delivered to, inspected, and accepted by, ANL.

All the natural UO_2 pellets have been fabricated, tested, and inspected. All UF_6 for enriched UO_2 pellets has been converted to oxide, and blends of enriched UO_2 with the required concentration of rare earth oxides have been completed at the Chemical Operations of the United Nuclear Fuels Division.

Facilities have been prepared for fuel loading, welding, handling, and inspection of the fuel rods. All Zircaloy-2 strip stock for fuel can and spacer grids has been fabricated by Wah Chang Corporation under subcontract to United Nuclear Corporation. Forming and welding facilities are being prepared for the fabrication of fuel element cans and spacer grids.

3. Plutonium Recycle Control Rods

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

The boron-stainless steel is being fabricated by Carpenter Steel Company. This steel will be finished to strip stock $0.33 \times 28 \times 153 \text{ cm}$ ($0.130 \times 11 \times 60 \text{ in.}$).

All hafnium for the oscillator rod is being converted to plate stock by Wah Chang Corporation from material furnished by ANL.

The completed control rods and oscillator rod will be fabricated from boron-stainless steel and hafnium. Bids have been received from commercial fabricators and are being evaluated before a contract is awarded for this work.

4. Nondestructive Tests on Components of Plutonium Recycle Experiment

Thirty-seven Zircaloy-2 tubes, each 1 3 meters long of 1.08 cm OD x 0.068 cm wall, were inspected by pulsed, eddy-current, test equipment as a check on the quality of the tubing and the effectiveness of the vendor's non-destructive test. The maximum allowable defect in these tubes, according to the specification, is a 5% localized reduction in wall thickness. No defects of this size or larger were detected in any portion of any tube except for one pit so close to the end of one tube that nondestructive test equipment could not reasonably be expected to detect it. Shallow scratches probably created during the air-gage ID measurement were noted in some tubes. The tubing was inspected on a pneumatically driven conveyor that scans the tube longitudinally and rotates it for the next inspection pass. Twenty-one passes were used on each tube at a rate of ten passes per minute.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Core Physics

a. Burnup Calculations. Calculations are underway using CYCLE 2 with special dimensions.⁶ The special dimensions are as follows:

Number of energy groups	≤ 20 ;
Number of regions	≤ 25 ;
Number of materials	≤ 24 .

Volume-weighted averages are being used for calculating concentrations of Xe^{135} and Sm^{149} since the composite material 9 (mixture of Sm^{149} and Xe^{135}) must now be used in place of materials 1 (U^{235}), 3 (Sm^{149}), and 24 (Xe^{135}). U^{238} will be added as a material in the burn-up calculations and will be assigned identification number 6.

Debugging of the recompiled code should be finished shortly.

b. Streaming Effect. The effect on reactivity caused by stream from the beam tubes has been determined to be

$$\Delta k/k = -0.21\%.$$

The reactivity change was calculated using the REX Code and the 18-group cross-section set. A PERT calculation gave a similar result. This effect, which is over and above the straight density effect, was calculated using Behrens' theory.⁷ The smallness of the effect is ascribed to the water layer between the inner and outer beryllium reflectors.

c. Fluxes in the Exterior Water. Neutron fluxes (between the outer beryllium and the pressure vessel) have been redetermined using linear anisotropy and DSN. The attenuation of the fast neutrons determined by this method is the same as that previously obtained by Monte Carlo theory.

d. Angular Fluxes. Neutron fluxes as a function of position and radius have been determined. There is little anisotropy (except for gradient effects) in all fluxes except the highest energy groups. Thus tangential

⁶B. Toppel, R. Avery, and G. Fischer, Cycle Cost Codes for Fast Reactor Fuel Cycle Analysis and Related Costs and Evaluation, Am. Nuc. Soc., Vol. 5, No. 1, p. 92.

⁷D. J. Behrens, The Effect of Holes in a Reacting Material on the Passage of Neutrons, Proc. Phys. Soc. (London), Vol. 62: Series A, p. 607 (1949).

beam holes offer a substantial reduction over radial tubes (by about an order of magnitude) in the neutrons above 3 MeV only. This effect should be more substantial for gamma fluxes.

Thermal fluxes are isotropic, and collimated thermal beam currents are estimated at 2×10^{11} n/cm²sec.

2. Shielding and Preliminary Safety Analysis

a. Plume Doses. Whole-body, beta-ray and gamma-ray, plus thyroid doses have been computed for fission products in an elevated plume produced by forced ventilation of the AARR reactor building (see April 1964 Progress Report, ANL-6886).

Only the gases are significant for the external doses since the release from the core, building, and filtration system is 100, 0.25, and 0.005% of the core inventory during shutdown for the gases, halogens, and solids. Decay enroute, and the dose from the daughter products of the gases, are nearly compensatory at the maximum dosage point and are both neglected for the external doses. Iodine precursors are not included for the thyroid dose since their effect on the total dose is small.

Results indicate ventilation rates should be large enough to ensure building inleakage and controlled release through the filtration system but should be less than 10 cfs. The maximum long-period doses for a 76-meter stack height, a 10-cfs flow rate, perfect mixing flow, a 2-meter/sec wind speed, and category E meteorological conditions are as follows:

Whole-body Plume Dose, Roentgens

<u>Beta-ray</u>	<u>Gamma-ray</u>	<u>Thyroid Dose, Rads</u>
5 (2-5 km)	13 (1-2 km)	30 (2-5 km)

The thyroid dose would be a maximum of 30% more for slug flow through the reactor building, and the other doses would be similarly increased.

b. Argon-41. The subpile room and experimental beam tubes are sources for argon-41. Assuming an external beam of 1% of the beam tube neutron current,⁸ the beam tube contributes about one-half of the argon-41. Biological dose limits determine minimum local ventilation rates of 10 cfm/experiment around the pile face; but when discharged from the stack, the concentration is not expected to exceed 10^{-16} c/ℓ at 1 km from the stack.

3. Fuel and Core Design

The fuel development program has progressed to the stage where samples are being processed. Fuel samples for evaluation of current

⁸D. Connor, Private Communication, ANL

manufacturing processes are being purchased from manufacturers currently producing military reactor fuel. Current fabrication processes and end products will be evaluated with regard to the AARR general requirement (i.e., 37 w/o UO_2 -SS, 40-mil plates with 5-mil clad) and will determine the realistic base from which the fuel development program must proceed. The testing of these samples will terminate this first phase of the program (i.e., Phase I, Evaluation of Existing Technology and Fabrication Methods).

Requests for advanced fuel development on cermet fuels have also been initiated. The effect of variation in intermediate stages of manufacture will be evaluated with regard to the end product, the ultimate aim being the production of a 37 w/o UO_2 -SS cermet fuel that can be mass-produced to exact dimensional tolerances, with homogeneity of the dispersion, and negligible fracturing, stringering, or clumping of the oxide. All data from Phase I will be fed into Phase II (Development of Advanced Technology) of the work. The end product of this study will be the manufacture of specimens for irradiation, which is scheduled to begin in November 1964.

Developmental work has also been initiated on assembly techniques of manufacturing the rhomboidal fuel assembly.

Stainless steel dummy fuel assemblies are being life-tested under the hydraulic flow study program. They are being tested for erosion and fatigue at elevated temperatures in high-velocity water.

4. Reactor Component Design Studies

A research and development contract has been let to provide design and design values for reactor components and the reactor system components. This contract has six major sections, each section representing a logical subdivision of the reactor design. These subdivisions are as follows:

- a. Prototype design and testing of beam tube assemblies.
- b. Reflector design and analysis.
- c. Nuclear instrumentation study.
- d. Process control stability study.
- e. Fuel transfer system design study.
- f. Control rod drive mechanism concept review.

5. Critical Experiment

Preparations for the AARR Critical Experiment continued as expected during May with the exception of hafnium for the control blades. The delay encountered in hafnium fabrication will crowd sequential items and prevent leveling-off of the work load, although it has not yet become a limiting item.

The delay started with the discovery that part of the hafnium supplied to a subcontractor was not satisfactory for rolling. Although a satisfactory substitution has been made, it involved substantial delay and increased cost.

Fuel processing appears to be on schedule, and delivery of bare uranium foils to Argonne should start during June. Stainless steel envelopes are to be welded over each piece of uranium by ANL's Central Shops, using an electron-beam welder to seal the edges. Depending on progress with envelopment, it may be possible to load fuel into the critical experiment during July and, consequently, other preparations are being moved up to accommodate a July startup (rather than August).

Current activities are installation of neutron flux detectors; submission of a preliminary statement of Operating Limits for the critical experiment (a new item); preparation of figures and forms for operating procedures; fabrication of remaining components for fuel assemblies, control blades, and followers; and preparation of a retainer for the beryllium reflector.

Discussion of location of beam tubes in the beryllium reflector and measurements required during the critical experiment have indicated the following course of action. Installation will not be necessary for several months. Beam tubes will be square and will be placed in only a limited number of perpendicular and tangential locations, relative to the fuel zone, which involve little or no refabrication of beryllium to accommodate these facilities.

Measurements in the beam tubes will be supplemented (if this proves to be feasible) by measurements in other research facilities. The motivation is that many of the desired measurements are at the limit of the capabilities of the critical experiment. Heat generation measurements and neutron spectrum distribution measurements in beam tubes require such high flux levels as to interfere with access to the facility for some days thereafter.

B. Magnetohydrodynamics (MHD)

1. MHD Power Generation - Jet Pump Studies

Studies were carried out to determine the comparative characteristics of several sizes of the compound and converging-diverging nozzles in pumping water in the injector loop. Preliminary examination of collected data confirmed the calculations made when the nozzle was designed. As expected, the annular compound nozzles show a linear characteristic at the same time the central nozzle asymptotically approaches a limiting velocity. This effect is caused by the orifice restrictions feeding the vapor to the outer annuli, which in turn forces the nozzles to operate over a range of linear response. The center-nozzle steam velocity is in the vicinity of 2300 ft/sec for 100-psi steam-feed pressure.

The water-pumping velocities were calculated from the measured volume flow-rate, corrected to the cross section of the narrow portion of the combiner. These figures are based upon the assumption that the flow is all uniformly single phase, but actually a two-phase situation exists. The extent of the two phases is not known since densities and pressure at the throat are not determined. Hence, the velocities may be a considerable order of magnitude greater than the calculated figures. These values will be measurable when the nozzle is operated with a liquid metal, since the velocity will be a direct function related to the potential generated in a transverse magnetic field. When the nozzle was operating with water, the pumping capability of the compound nozzle was considerably better than that of the converging-diverging nozzle. The control and response were more stable over a wide range, and actual velocities reached were higher for any given set of conditions. Part of this superior performance of the compound nozzle was attributable to a higher steam discharge rate for a given pressure. However, even when the pumping rate was normalized to compensate for the discharge quantity, the compound nozzle showed better pumping characteristics. At the limiting velocity, both the compound and converging-diverging nozzles began to pump equally well. This is probably due to the predominance of slip losses, which became similar for both nozzles.

The response to back pressure was also studied in both types of nozzles, since loading by the generator will produce such a force in the linear generator model. Again, the compound nozzle showed excellent stability with flow velocity dropping off in inverse ratio to back pressure. On the other hand, the converging-diverging nozzle tended to be very critical and tended to maintain a constant value of velocity over a range, with abrupt cessation of operation when the back pressure range was exceeded. This latter effect is probably caused by the establishment of a sharp condensing shock, which establishes a set of stable conditions, permitting the jet to pump more effectively as the back pressure is increased. When the back pressure exceeds this value, the condensing shock breaks down and the vapor tends to slip past the liquid, causing the stability conditions to break down sharply.

The experimental nozzles and two commercially available units were compared when the system was operated at 90-psi steam-feed pressure. The comparisons are not precise since feedwater temperatures for the recirculating system tend to be somewhat higher than desired. The comparison data is tabulated in Table XXXIX.

Table XXXIX. Steam Nozzle Operation with 90-psi Steam

<u>Nozzle Type</u>	<u>Velocity, fps</u>	<u>Nozzle Type</u>	<u>Velocity, fps</u>
Penberthy No. 23A	23.1	Compound	88
Simple, single stage	68	Penberthy No. 3A	22.5
Converging-diverging	74		

The velocities, again, are based upon single-phase flow. The comparison shows that higher velocities have been reached with the compound nozzle than with other types. This is important since the voltage is dependent upon the flow velocity.

2. MHD AC Generator Flashing Cycle

The apparatus for the AC Pulsed Flow MHD experiment has been assembled. A large nitrogen supply has been obtained so that longer runs can be made, and the generator is being instrumented for power runs. The instrumentation includes a pressure transducer which will be used to measure the pressure of the driving gas behind each slug of fluid.

The continuity of the liquid slug will be determined from the voltage pulses from the generator. Magnetic field intensity will be measured with a Hall device. The output from these instruments will be recorded on an oscillograph. The generator will be operated under various conditions of fluid pressure, drive-gas pressure, field intensity, and loading. From this information, the stability of the liquid-metal slug can be evaluated under various conditions.

C. Regenerative EMF Cells

1. Bimetallic Cell Studies

a. Spectrophotometric Studies of Li_2Te and Cs_2Te in Molten Salts.

The absorption spectrum of Li_2Te in LiCl-LiF at 525°C has been reported previously (see Progress Report for February 1964, ANL-6860, p. 95). The spectrum was characterized by an absorption peak at $469\text{ m}\mu$, an absorption minimum at $385\text{ m}\mu$, and an intense absorption band with its long wavelength edge at $377\text{ m}\mu$.

In recent experiments, the absorption of Li_2Te in LiCl-KCl (50 m/o LiCl) was studied. The spectrum at 500°C was found to be nearly the same as that of Li_2Te in LiCl-LiF ; however, the absorption maximum was shifted slightly and occurred at $485\text{ m}\mu$.

The absorption spectrum of Li_2Te in CsCl at 700°C was also determined. Two absorption peaks were found, one at about $465\text{ m}\mu$, and the other at about $635\text{ m}\mu$. A charge-transfer band with its long wavelength edge located at about $300\text{ m}\mu$ was also observed.

The spectrum of Cs_2Te in CsCl was also measured. At 700°C , the absorption spectrum exhibited two maxima, a strong absorption peak at $635\text{ m}\mu$, and a weak absorption peak at $465\text{ m}\mu$. A charge-transfer band with its long wavelength edge located at $340\text{ m}\mu$ was also observed.

2. Lithium Hydride Cell

The standard emf of the lithium hydride cell

has been determined to be 0.2908 Volt at 540°C. The effect of the solubility of LiH in lithium was ignored, because the total emf error introduced (assuming an ideal solution) would be less than 2 mV. The calculated value for the Gibbs free energy of formation of LiH(s) was calculated to be -16,400 cal/mole at 298°K. This is in good agreement with literature values at the same temperature, namely, -16,450 cal/mole⁹ and -16,800 cal/mole.¹⁰

3. Phase Diagram Studies of LiH-NaCl System

The phase diagram of the LiH-NaCl system has been determined by thermal analysis. The system was found to be a simple eutectic melting at 565.7°C and occurring at 55.4 m/o LiH. Both the sodium chloride and lithium hydride appear to remain un-ionized in the system.

⁹T. R. P. Gibb and C. E. Messer, USAEC Report NYO-3957, August 1957.

¹⁰S. R. Gunn and L. G. Green, J. Am. Chem. Soc., 80, 4782 (1958).

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal-Water Reactions

a. Studies of Aluminum-Water Reactions. The aluminum-water reaction has been studied by several different methods. The important results of these studies are summarized here because, for this field in particular, fragmentary results are unusually difficult to tie together. Studies of the aluminum-water (or steam) reaction can be divided into four groups: 1) experimental determinations of the isothermal rate law and rate constants, 2) nonisothermal studies of the reactions of small metal particles, 3) in-pile studies in TREAT, and 4) calculational studies, including data correlation and accident analysis.

(i) Experimental Determinations of Isothermal Rate Law and Rate Constants. Isothermal studies have been performed by two methods. The pressure pulse method (see ANL-6413, p. 179) was satisfactory only over the range 800 to 1200°C. The levitation melting method was satisfactory over the range 1200 to 1750°C (ANL-6648, p. 196). In the pressure pulse method, the molten metal is contained in a crucible in an evacuated reaction cell and is brought to the desired temperature by induction heating. Electronically-controlled valves are operated in the proper sequence to contact the molten metal with steam at 1 atm for a specified period of time. Extent of metal-water reaction is determined by analysis of hydrogen in the exit gas. The results showed that the aluminum reaction with steam at 1 atm resulted in the formation of a very protective oxide film. The reaction rate can be described by a cubic rate law at 800, 1000, and 1200°C for reaction times of 1, 10, 100, and 1000 sec.

The levitation melting method involves the simultaneous heating and levitation of a nominal $\frac{1}{4}$ -in.-diameter molten metal pellet in a radio-frequency field while flowing steam is passed over the sample. Pellet temperature is measured and controlled by a two-color optical pyrometer. The quantity of reaction is determined gravimetrically by selectively dissolving unreacted aluminum and leaving the oxide intact. Pellets of pure aluminum, an alloy containing 81 w/o Al, 17 w/o U, and 2 w/o Ni (SL-1 composition), and aluminum alloy 6061 (1 w/o Mg, 0.6 w/o Si, 0.25 w/o Ce, 0.25 w/o Cr, remainder aluminum) were exposed to steam for periods from 5 to 100 minutes at temperatures from 1200 to 1750°C. These heating periods included the times required to bring the specimens to the test temperature.

The results at 1200 and 1300°C for aluminum and the two alloys studied could be described by a cubic rate law. However, the scatter of the data was generally greater than the scatter of pressure pulse data at

1200°C. The extent of reaction at 1200°C by the pressure pulse method was approximately twice the extent in the levitation studies. The greater reaction obtained by the pressure-pulse method very likely resulted from the diffusion of steam through the slightly porous alumina crucibles used to contain the molten aluminum, which allowed reaction to occur over an increased surface. The cubic rate law was fitted to the levitation data, and the following equation was obtained:

$$W^3 = 4.0 \times 10^9 t \left[\exp \left(\frac{-73,500}{RT} \right) \right], \quad (8)$$

where

W = metal reacted, mg Al/cm²;

t = time, minutes;

R = gas constant, 1.987 cal/(mole)(°K);

T = temperature, °K.

The activation energy, 73,500 cal/mole, found experimentally is approximately the same as the values reported for the heat of vaporization of aluminum. A similar coincidence between the activation energy for oxidation and the heat of sublimation of magnesium was taken as evidence for a vaporization-controlled reaction.¹¹

Equation (8) describes, within a factor of two, the reaction of aluminum and the two alloys of aluminum with steam at 1 atm over the range 800 to 1300°C. Equation (8) also describes the reaction of the aluminum-uranium-nickel alloy (SL-1 composition) at 1400, 1500, and 1600°C, although the values of the experimental points at 1600°C are about 50% greater than the calculated values. Pure aluminum appears to follow a linear rate law at 1400, 1500, and 1600°C. It is also possible, however, to fit the data satisfactorily to a curve that follows a cubic rate law initially and then undergoes a smooth transition to a linear rate law after 2.3 mg Al/cm² has reacted. The linear portion of the curve is described by the following equation:

$$W = 2.5 \times 10^8 t \left[\exp \left(\frac{-73,500}{RT} \right) \right], \quad (9)$$

where

$W > 2.3 \text{ mg Al/cm}^2$.

¹¹Gibbs, D. S., and Svec, H. J., Kinetics of the Reaction between Magnesium and Water Vapor, ISC-779, Iowa State College, 1956.

Values of the extent of reaction of 6061 aluminum alloy were very scattered and were generally greater at 1400, 1500, and 1600°C than those given by Eq. (2). This greater reaction was probably due to the disruption of the protective oxide film by vaporizing magnesium.

Pure aluminum and the two alloys studied ignited immediately in steam (and also in air) when the metal temperature reached 1750°C. The burning reaction that followed was evidently a combustion process in which aluminum was first vaporized (vapor pressure ~ 7 mm at 1750°C) and then reacted as a vapor. Very fine aluminum oxide powder was formed during the burning. Pellets that had been ignited in air or steam, and had been subsequently dropped into liquid water, continued to burn with increased vigor under the water surface and later at the water-air interface. The consumption time of pellets was measured as a function of diameter; the surface burning rate was found to be independent of diameter. The rate was constant at $0.96 \text{ g Al}/(\text{cm}^2)(\text{min})$. Pellet consumption, however, was not complete. An oxide shell of irregular thickness formed around the pellets during the latter stages of burning, and the equivalent of a 2.3-mm-diameter sphere of aluminum remained unreacted after the burning had ceased.

Delayed ignitions occurred at temperatures as low as 1600°C (15-min delay with pure aluminum). These delays were very likely due to the ignition of Al_2O , a suboxide which has been reported to form when aluminum and Al_2O_3 are together at high temperatures. Thermodynamic estimates indicate that Al_2O may have a vapor pressure as high as 7 mm at 1600°C. The delay in ignition, therefore, is probably related to the time required to generate a significant quantity of Al_2O_3 .

(ii) Nonisothermal Studies of Reactions of Small Metal Particles. Nonisothermal studies of small particles have included experiments using the laser-beam heating method. In this study, small sections of aluminum foil under water are heated in about one msec by a focused light beam from a ruby laser. Results with aluminum are listed in Table XL. The small sections of foil are generally converted to molten spheres during the heating process; however, in one case, a violent breakup was seen on high-speed motion pictures, and in two runs with thick foils, the specimens were not fully melted. The extent of metal-water reaction was determined by measuring the hydrogen generated using microchemical analysis. The small foil (see Table XL) reacted from 7 to 11%, the extent of reaction being independent of the laser energy. High-speed motion pictures showed the growth and collapse of the steam-hydrogen bubble surrounding each particle; vapor phase burning was observed in only one run in which a $2 \times 2 \times 0.02$ -mm foil was used.

Table XL. Extent of Reaction for Aluminum Heated under Water by a Light Beam from a Ruby Laser

Foil Size (mm)	Equivalent Sphere Di- ameter (μ)	Laser Energy (Joules)	% Reaction
1 x 1 x 0.02	370	10	7.3
		15	10.0
		18	6.8
		25	10.5
		29	7.4
		42	11.4
		48	9.5
2 x 2 x 0.02	580	44	28.2 ^a
		50	14.9
2 x 2 x 0.13	980	49	0 ^b
		49	0 ^b

^aFoil broke up violently.

^bSpecimens may not have been fully melted.

Attempts to correlate the results of isothermal rate studies with those obtained by the laser method were made using a computer code developed in connection with the zirconium-water reaction (see ANL-6548). The cubic-rate-law equation (Eq. 8) was used along with a simple heat-balance equation to generate the transient temperature and extent of reaction of small molten particles that were suddenly exposed to water. The results of the calculations are given in Table XLI. The 7 to 11% reaction obtained experimentally with a 370- μ particle by the laser method is not predicted by the calculations. It was concluded, therefore, that the reaction of small spheres under relatively violent conditions differs in character from the reaction obtained under carefully controlled conditions in isothermal experiments.

Table XLI. Computed Reaction of Small Spheres of Molten Aluminum with Water^a

Diameter (μ)	Percent Metal-Water Reaction			
	Initial Temperature ($^{\circ}$ C)			
	1400	1500	1600	1700
1050	0.20	0.30	-	0.59
262	0.37	0.59	0.92	1.31
66	-	1.16	-	3.66

^aBased on cubic rate law, Eq. (8).

(iii) In-pile Studies in TREAT. Studies of the reaction of water with representative aluminum-base fuel materials in TREAT have

WATER TEMP (°C)	SYSTEM PRESSURE (psi)	FUEL SPECIMEN (Plates)
25	20 ^a	○ { SPERT 77 w/o Al, 23 w/o U, Al CLAD
285	1000	● {
25	1000 ^a	△ {
25	20 ^a	□ { SL-1 81 w/o Al, 17 w/o U, 2 w/o Ni, UNCLAD
285	1000	■ {

Figure 32. Aluminum-Uranium Alloy Plate Meltdown Experiments in TREAT

of powdery alumina, which is indicative of vapor phase burning. The results with heated and pressurized water showed that very extensive reaction (75 to 90%) occurred when the estimated peak temperature of the fuel plate exceeded 1200 to 1400°C. The residue from these runs also suggested that burning had occurred. The extent of reaction in room-temperature water which was pressurized to 1000 psi with helium was intermediate (38% reaction); that is, the extent of reaction was between that found in room-temperature water at 20 psi (10 to 20% reaction) and that found in water that was heated and pressurized (75 to 90% reaction). One experiment was completed with preirradiated SL-1 fuel material. Although 2.2 a/o of the heavy atoms present in the fuel had been fissioned, preirradiation had no apparent effect on the appearance of the residue or the extent of metal-water reaction.

The extreme sensitivity to pressure of the aluminum-water reaction was unexpected, since other experiments had shown that the zirconium and stainless steel-water reactions were relatively unaffected by large pressure changes. This had been demonstrated by the condenser discharge experiment in which small metal wires were heated and melted in the presence of water by a surge current from a bank of condensers. Attempts to conduct the condenser discharge experiment with aluminum wires failed because the wires were not heated uniformly, as was revealed

shown the extent of reaction to be considerably greater than that predicted from the isothermal studies. TREAT results summarized in Figure 32 indicate that 15% reaction occurs in transient experiments in which the peak temperatures of the specimens submerged in room-temperature water are estimated to be 1200 to 1400°C. In the TREAT experiments, two types of fuel specimens, unclad SL-1 plate material and clad SPERT 1-D plates, were placed in small, high-pressure autoclaves and located at the center of TREAT. Most of the experiments were performed with the specimens immersed in room-temperature water with a 20-psi helium overpressure; one experiment was performed using room-temperature water and a 1000-psi helium overpressure. Residues from high-energy runs indicated the presence

by high-speed motion pictures. Instead, the wires broke, and the electrical energy was dissipated in arcing. Recent experiments have been undertaken, however, using aluminum wires encased in nickel tubing and the assembly drawn to an outside diameter of 29 mils. This coaxial assembly was heated to temperatures between 1400 and 2000°C by discharging condensers through the assembly. Table XLII gives the preliminary results of this study. The data indicate that steam at high pressure greatly increases the extent of reaction, but that inert gas suppresses the reaction.

Table XLII. Extent of Metal-Water Reaction for Composite Aluminum-Nickel Wires Heated under Water by Discharging Condensers

Water Temperature (°C)	Pressure (psig)	Pressurizing Gas	Calc. Metal Temperature (°C)	Mean Particle Size (mils)	Percent Al-H ₂ O Reaction
22	2000	He	1970	-	<0.4
22	1500	He	2050	1.0	<0.4
22	1000	He	1930	3.2	<0.4
22	500	He	1960	2.8	0.8
22	0	He	2000	-	2.7
242	500	Steam	1440	4.5	20.3
285	2000	Steam	1820	5.8	58.2

(iv) Calculational Studies, Including Data Correlation and Accident Analysis. In an attempt to clarify the effect of pressure on the aluminum-water reaction, and for other purposes (such as simulation of environment in a loss-of-coolant accident), a high-temperature, high-pressure furnace is being constructed. It is expected that steam pressures up to 1000 psi and temperatures up to 1900°C can be reached. Isothermal reaction rates of aluminum-water reactions and ignition temperatures of aluminum and aluminum alloys will then be determined as a function of pressure as well as temperature.

Development work is also in progress on a method whereby meltdown and fuel plate breakup in TREAT experiments can be observed by means of high-speed motion pictures. Motion pictures of TREAT experiments have been taken in the past, but not of experiments using water. It is expected that the special problems created by the presence of water can be solved and that the motion pictures will provide needed insight into the mechanism and rate of fuel plate breakup and reaction.

At present, the extent of the aluminum-water (or steam) reaction at high temperatures must be viewed as being very dependent upon environmental conditions, such as pressure, water temperature, particle size, and the presence of turbulence. Thus, in steam at 1 atm and in the absence of turbulence, aluminum forms a very protective oxide up to temperatures of 1750°C, at which temperature ignition occurs. Under more violent conditions, such as those prevailing in laser-heating experiments or

in TREAT experiments, the extent of reaction is considerably greater at temperatures below 1750°C. It is not presently known whether this increased reaction under turbulent conditions results from repeated fracturing of the oxide film or by sporadic vapor burning. The failure to observe a very rapid pressure rise in TREAT experiments indicates that the reaction under turbulent conditions is not extremely rapid. The extent of reaction is increased further when the reaction takes place in heated water at a high overpressure of steam, suggesting that either the reaction rate is greatly increased or the ignition temperature is lowered. The sharp energy threshold for reaction in TREAT experiments (see Figure 32) suggests that the lowering of the ignition temperature is the most likely cause of the increased reaction, although it is difficult to find a mechanistic explanation for this behavior.

TREAT results have been shown to be consistent with the analyses of debris from the SL-1 accident and the final destructive test of the SPERT 1-D core. In making the comparisons, the reactor cores were segmented mathematically into regions according to the maximum temperature reached. The quantity of metal in each region was then multiplied by the percent reaction, based on the room-temperature water data of Figure 32. The metal reacted in each region of the core was then summed to yield the overall reaction. These values were then compared with the extent of metal-water reaction found by the chemical analysis of α -alumina in the reactor debris. The results were as follows:

	Energy Generated by Metal-Water Reaction	
	Observed	Calculated
SL-1 Accident	24 $\pm$ 10 MW-sec	26 MW-sec
SPERT Experiment	3.5 MW-sec	2.0 MW-sec

b. Calculation of Pressure History of a Reactor during an Excursion. Calculations of transient heat transfer and pressure buildup occurring during violent nuclear excursions were continued. The present effort is directed toward the mathematical simulation of the observations recorded during the final destructive test of the SPERT 1-D core. The calculational method involves the solution of two sets of simultaneous equations. Solution of the first set of equations yields values for the thickness of the steam film and the pressure at the core surface during the time (2 msec) of sonic loading of the water column above the core. The solution of the second set of equations yields values for the steam film thickness and pressure during the time immediately following the sonic loading period, when inertial considerations replace those of acoustic constraint. The results of calculations for the first pressure pulse in the SPERT destructive test were reported previously (see Progress Report for April 1964, ANL-6885, p. 70).

Calculations intended to simulate the second and largest pressure pulse in the SPERT test have been undertaken. Extensive breakup of the molten region of the core occurred during the second pressure transient and resulted in a greatly increased surface area. Calculations are being made first for the case in which the steam generated during the first pulse and the time interval between the first and second pulses are ignored.

Another calculation is being made in which the steam generated during the first pulse and during the time interval between the first and second pulses is considered to be a compressible gas.

c. Studies in TREAT of Uranium Dioxide-Core, Zircaloy-2-Clad Fuel Pins. A fairly substantial body of information has been obtained on metal-water reactions in TREAT with UO_2 -core, type 304 stainless steel-clad fuel pins and with mixed oxide-core, Zircaloy-2-clad fuel pins. However, more information is needed concerning the reaction of UO_2 -core, Zircaloy-2-clad fuel pins with water when exposed to transients in TREAT. An experiment with a UO_2 -core, Zircaloy-2 clad fuel pin has been described previously (see Progress Report for March 1964, ANL-6880, p. 73). The conditions and results of a second experiment in which a single fuel pin was exposed to a very energetic transient in TREAT while submerged in room-temperature water are given in the following table:

<u>Run No.</u>	<u>CEN-175</u>
<u>Reactor Characteristics</u>	
Integrated Power, MW-sec	505
Peak Power, MW	2550
Reactor Period, msec	52
Energy Input, cal/g	450
<u>Fuel-Pin Characteristics</u>	
Core Material, % UO_2	100
Enrichment, % U^{235}	11.2
Cladding Metal	Zircaloy-2
<u>Results</u>	
Percent Metal Reacted with Water	63
Final Geometry of Fuel Pin	fine particles
Peak Pressure, psi	180

The extent of metal-water reaction obtained in Run CEN-175 was consistent with the results obtained in the first run (CEN-173). In Run CEN-173, 79.8% reaction occurred for an energy input of 694 cal/g.

2. Metal Oxidation-Ignition Studies

Experimental studies of the thermal conductivity of metal powders have been completed. A simple theory, similar to that developed by Deissler

and Eian,¹² has been developed to describe the results obtained. The theory assumes that spherical particles are arranged in an orthorhombic structure which has a solid volume fraction of 60.5%. The theory applies to particle-gas systems in which the Knudsen number (ratio of mean free path to particle diameter) is less than 7×10^{-4} . The effective powder conductivity is a function only of the conductivities of the solid and the gas. The theory shows good agreement with experimental results for unoxidized metal powders. Means of including the case of metal powders, each particle of which has an oxide coating, are being studied.

B. Fast Reactor Safety Studies

1. Integral Sodium Loop

The integral sodium loop has been tested at the experimental pre-transient operating temperature of 260°C, for several hours of running time. All operating and instrumentation circuits performed perfectly, with all interlocks and safety circuits sequencing and in satisfactory order. The movements of controllers and recorders were secured, and the loop assembly was packed for shipment to the TREAT facility. An experimental transient test using an assembly in which an internal thermocouple has been installed in a fuel pin is scheduled for next month.

The remote assembly-disassembly apparatus to be used with the small loop was tested extensively and the required techniques for loading and unloading the loop, while filled with molten sodium, were developed.

2. Stand-off Pressure Transducers

The development of a prototype penetration pressure tap for use with the large sodium loop has been completed and is ready for installation in the test-section mockup. The mockup will be filled with sodium and operated at temperatures up to 500°C during the test.

Static calibration tests of the variable-length stand-off pressure transducer assembly, constructed for use in the frequency-response experiments, revealed that the threaded-nut and ferrule type of tubing connector is unsuitable for use in liquid sodium or NaK-filled systems which are cycled over wide pressure ranges. The variable-length standoff assembly was reconstructed using a fully-welded technique.

3. Large TREAT Loop

The contract for building the large cask to be used in shipping fueled portions of the TREAT loop has been awarded. The estimated date of delivery is the latter part of June 1964.

¹²Deissler, R. G., and Eian, C. S., NACA-RM-E52-CO5.

Portions of the cold trap assembly have been received from the shop and are being assembled before shipment to Idaho.

After a sample has been subjected to an excursion in the TREAT reactor, the assembly is cooled and the sodium solidified. The solid sodium resists the removal of the experimental section of the loop. The forces developed on the upper flow tube bellows by frozen sodium during header and pressure container separation have been evaluated. The study indicates that if the level of the frozen sodium is more than 2 in. above the joint, the bellows will be held immobile during separation. A restraining force will not be present if sodium is frozen to a level below the joint, thus permitting the bellows to expand to its natural length as the disassembly is carried out. Sodium will have to be frozen "in situ"; however, arrangements can be made to lower the level sufficiently to accommodate the unloading procedure.

VI. PUBLICATIONS

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