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REACTOR DEVELOPMENT PROGRAM
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FOREWORD

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

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

The last six reports issued
in this series are:

September 1963	ANL-6784
October 1963	ANL-6801
November 1963	ANL-6808
December 1963	ANL-6810
January 1964	ANL-6840
February 1964	ANL-6860

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I. BOILING WATER REACTORS

A. BORAX-V

1. Operations and Experiments

Loading of the peripheral superheater core was completed and zero-power critical experiments were begun this month. Prior to loading fuel into the reactor, the control rods and drives were test-operated and performed satisfactorily. The source and nuclear instrument chambers were positioned in the reactor vessel to measure neutron multiplication changes during loading.

To determine the minimum critical mass with peripheral superheater fuel, sixteen available fuel assemblies were loaded in a square array. The assembly was found to be subcritical, and the fuel assemblies were unloaded.

Boiling fuel assemblies, containing 37 fuel rods (4.95% enriched), and twelve flow rods were then loaded to find a minimum critical mass. Fourteen assemblies were required. The loading was then modified to obtain a rectangular slab core with partially loaded fuel assemblies on one side, as shown in Figure 1. Reactivity measurements with two arrangements were made. The core having twelve complete assemblies and four assemblies with two rows of fuel rods and flow rods was subcritical. With three rows of fuel and flow rods, however, the reactor achieved criticality with all control rods nearly fully withdrawn.

To determine the effect of poison rods on a square core containing sixteen fully loaded boiling fuel assemblies, measurements of three arrangements were performed. First, no poison rods were used; then eight poison rods were substituted for flow rods, finally, eight poison rods were substituted for fuel rods. Flux-wire irradiations were made with the minimum critical core and the 16-assembly core to determine axial flux distribution.

Several poison rod locations were tried to obtain the maximum available excess reactivity without exceeding the requirement for the reactor to be subcritical with any single control rod withdrawn. For the 60-assembly loading, 40 boron-stainless steel poison rods in place of fuel rods are required to prevent single-control-rod criticality.

The reactivity effect of draining the superheater was found to be about -1.5% at room temperature, determined from the change in critical position with all control rods banked.

Flux-wire irradiations of the final poisoned core were made at room temperature to determine the fine radial neutron flux distribution and coarse radial and axial flux distribution.

Figure 1. Minimum Critical Slab with Boiling Fuel, 12 Flow Rods per Assembly, BORAX-V

2. Reactor Physics Calculations

Several additional problems have been run by means of RE-122 and PDQ and the 3-group No. 780 cross-section set (see Progress Report for January 1964, ANL-6840, p. 2). The distributed 12-flow-rods-per-boiler-assembly case (previously reported in ANL-6840, p. 4, as configuration III) was used in all the work reported here. A slight modification was found to be necessary in the hot (489°F) problems, owing to an error in atom density in the original MUFT calculation of the fast constants. Corrected values of these, together with the results of several other problems are outlined in Table I.

Table I. Calculated Core Characteristics (Superheater Cores for BORAX-V)

Temp (°F)	Steam-Void % of Boiling Water Volume	Superheater Fuel Loading (g U ²³⁵ /Assembly)	Superheater Condition	Superheater Power Fraction	Core k _{eff}	Core Available Excess Reactivity (%)	Superheater Flooding Reactivity Worth (%)
Central Superheater							
489 ⁽¹⁾	0	430	Flooded	0.223	1.126	11.2	1.5
489 ⁽¹⁾	0	430	Dry	0.157	1.108	9.7	
489	10	430	Dry	0.168	1.082	7.6	
489 ⁽¹⁾	0	680	Flooded	0.314	1.186	15.7	3.1
489 ⁽¹⁾	0	680	Dry	0.211	1.144	12.6	
489	10	680	Dry	0.225	1.121	10.8	
Peripheral Superheater							
489 ⁽¹⁾	0	680	Flooded	0.160	1.188	15.9	0.8
489 ⁽¹⁾	0	680	Dry	0.127	1.178	15.1	
489	10	680	Dry	0.139	1.147	12.8	
489	10	430	Dry	0.106	1.140	12.3	
70 ⁽²⁾	0	680	Flooded	0.142	1.273	21.4	
489 ⁽²⁾	0	680	Dry	0.127	1.190	16.0	
489 ⁽²⁾	10	680	Dry	0.138	1.159	13.7	
489 ⁽³⁾	0	680	Dry	0.137	1.153	13.3	
489 ⁽⁴⁾	0	680	Dry	0.138	1.152	13.2	
489 ⁽⁵⁾	10	680	Dry	0.146	1.130	11.5	
489 ⁽⁶⁾	10	680	Dry	0.149	1.123	11.0	
489 ⁽⁷⁾	10	680	Dry	0.157	1.108	9.8	
70 ⁽⁸⁾	0	680	Flooded	0.145	1.167	14.3	
70 ⁽⁹⁾	0	680	Flooded	0.144	1.173	14.7	
70 ⁽¹⁰⁾	0	680	Flooded	0.139	1.201	16.7	

(1) Values revised from those previously reported in Progress Report for January 1964, ANL-6840, p. 4.

(2) A "spiked" boiler region, 7.5 cm thick, was added just inside the peripheral superheater zone. This region had an effective enrichment of 7.5%.

(3) Includes a 5-cm-radius central zone with 10% voids.

(4) Includes a 10-cm-radius central zone with 10% voids.

(5-7) A study of power control with central control rod. A uniform poison occupies a region, 12 cm in radius, in center of boiler region.

Σ_a thermal in center increased by: 5 - 0.010; 6 - 0.015; 7 - 0.030.

(8) No spiked assemblies.

(9) Three corner assemblies in each quadrant spiked to equivalent of 7.5% enriched.

(10) Three corner assemblies plus outer row of boiler assemblies spiked to equivalent of 7.5% enriched.

A test of the No. 780 cross-section set was made upon loading the core with the minimum number of boiler assemblies, containing 12 flow rods per assembly, to achieve a critical configuration. The equivalent of approximately 14 boiling fuel assemblies was arranged in a rectangular slab by partially loading four assemblies, as shown in Figure 1. An axial flux traverse was obtained with enriched U²³⁵ alloy wires in the three locations shown in Figure 1 as W-1, W-2, and W-3. Table II presents the buckling values derived from the results.

Table II. Bucklings in Minimum Critical Slab, BORAX-V

Flux Wire No.	B _Z ² , cm ⁻²
W-1	0.00158
W-2	0.00155
W-3	0.00155

An approximation of the transverse buckling gives a value of 0.00756 cm^{-2} , or a total buckling of $B^2 = 0.00756 + 0.00155 = 0.00911 \text{ cm}^{-2}$. Fundamental-mode calculations infer that a critical buckling for the cross sections used would be 0.00968 cm^{-2} . This implies that the calculated k_{eff} for the minimum critical slab is within about 2%.

Similar calculations for a minimum critical assembly composed of peripheral superheater (PSH) fuel assemblies did not agree very well with experiment. From earlier predictions,¹ the area of a cylindrical core composed of 680-g U^{235} PSH elements suggested that nine flooded assemblies would be critical. The experiment indicated that a 4×4 array of 16 assemblies was subcritical. One explanation of this discrepancy is that the calculations overlooked the fact that the static steam-insulating gaps did not flood. Also, some question exists as to the validity of MUFT calculations of fast neutron absorption in stainless steel. Calculations of new constants, using the GAM and TEMPEST codes, are under way.

3. Radiolytic Gas Recombination

The presence of about 20-30 ppm of radiolytic oxygen in the steam from the boiler of an integral boiling-nuclear superheating reactor is believed to have a deleterious effect on both the oxidation rate and the chloride stress corrosion of austenitic stainless steels being used for cladding on some superheater fuel elements. Therefore, it is desirable to reduce significantly the oxygen content of the steam entering the superheater.

Experiments to determine the feasibility of recombining the radiolytic oxygen and hydrogen were undertaken by seeking a catalyst suitable for use in the internal steam dryer of BORAX-V. Criteria used for suitability were:

- a. catalyst will allow full flow of steam at $10,000 \text{ lb/hr/ft}^2$, with a pressure drop of 1 psig or less through a 4-in. bed depth;
- b. catalyst will effect recombination of a large percentage of the radiolytic gases;
- c. catalyst will retain physical integrity and activity for at least one year.

The catalysts are being tested under the following conditions:

Temperature:	489°F
Pressure:	600 psig
Oxygen concentration:	20-30 ppm
Hydrogen concentration:	Stoichiometric
Steam flow:	9000 lb/hr/ft
Bed depth:	4 in.

¹ Design and Hazards Summary Report Boiling Reactor Experiment V (BORAX-V), ANL-6302 (May 1961).

To date, the following six catalysts have been tested:

- a. low-surface-area copper mesh;
- b. platinized low-surface-area copper mesh;
- c. palladium oxide on activated alumina pellets (Girdler T-308);
- d. 0.7% palladium on low-surface-area alumina pellets (Girdler T-661);
- e. platinum oxide on activated alumina pellets (Girdler T-309);
- f. 0.5% palladium on activated alumina pellets (Girdler T-961).

No detectable recombination was observed except for the T-961 material which gave 22-27% recombination. This catalyst has been tested for over 30 hr and shows no sign of changing efficiency. The pressure drop across the 4-in. bed of T-961 is about 16 in. of water.

A sample of T-961 was removed after 7 hr of test and analyzed spectrographically. The catalyst was found to have taken up some traces of copper and zinc from the brass catalyst holder. Of further importance was the evidence that the γ - Al_2O_3 (gamma alumina) had hydrated to form α - $\text{Al}_2\text{O}(\text{OH})_2$, or Boehmite. The new samples of unused catalyst contained no crystalline palladium; however, after 7 hr of test the amorphous palladium had apparently changed to a crystalline form.

Testing of catalysts is continuing

II. LIQUID-METAL-COOLED REACTORS

A. General Fast Reactor Physics

1. ZPR-III

Experiments continued with Assembly 44, the engineering mockup of the RAPSODIE reactor. Mockups of the RAPSODIE control and safety rods were installed, and the final engineering configuration was attained. Measurements were then made of the worths of materials distributed in the core and in the transition zones between core and blanket (end gaps).

Prior to the construction of the mocked-up RAPSODIE B_4C control rods, the sodium-steel end gaps on the top and bottom axes of the core had been installed. The lower gap, 3 in. in axial length, was substituted for axial blanket; and the ZPR-III control rods in that half of the machine were then recalibrated. The upper gap was next constructed for a 6-in. length and finally extended to 7 in. The reactivity effects of the gap substitutions for axial blankets are given in Table III.

Table III. Worths of End Gaps in RAPSODIE Mockup

Zone	Composition	Relative Worth ^a (lh)
Axial Blanket	6.69 g/cc depleted U; 1.49 g/cc Steel; 0.378 g/cc Al; 1.04 g/cc Mo	0
3-in. Lower End Gap	3.06 g/cc Steel; 0.46 g/cc Na	-290
6-in. Upper End Gap	3.01 g/cc Steel; 0.48 g/cc Na	-381
7-in. Upper End Gap	3.02 g/cc Steel; 0.49 g/cc Na	-399

^aWorth of gap substituted for axial blanket.

The RAPSODIE reactor is composed of hexagonal elements, of which six at the radial edge of the core are designated for control and safety rods. Four rods, containing enriched B_4C , are safety rods; the other two, containing natural B_4C , are for control and reactivity shimming. In the Assembly 44 mockup, the six rods in the "up" position are represented by four "channels" loaded to give about the same total boron-10 concentration as in RAPSODIE. The channel positions, shown in Figure 2, have about the same average radii as the RAPSODIE rods.

Initial experiments were done in one channel. The radial blanket in the channel location was replaced with sodium-filled steel cans. In the portion of the channel below the intended location of the "active rod," some of the sodium was then replaced by steel to represent the rod stem. Finally, the proper mixture of enriched and natural boron carbide pieces was

installed in place of sodium in the "active" upper portion of the channel. This upper portion extended from 2 to 16 in. above the upper axial end of the core.

Figure 2. Core Configuration for RAPSODIE Engineering Mockup

In another channel, the "rod-plus-stem" was substituted directly for radial blanket and the worth measured. The remaining two "rods" were installed along with an addition of fuel at the core edge to maintain criticality.

The boron carbide portion of one of the "rods" was then moved in 3 in. and the reactivity effect determined. Subsequently, the other three "rods" were moved in 3 in., and this put the active portions of all the rods into a "banked" condition, 1 in. below the top of the core. With the necessary fuel adjustment for criticality, this completed the construction of the engineering mockup configuration, representing the RAPSODIE reactor during operation.

The resulting reactivity effects from the various substitutions and movements in the rod channels are compiled in Table IV.

Table IV. Worths of RAPSODIE Control-Safety Rod Mockups

Channel Loading (5.5-in. ² cross section)		
Axial Position (in. from core midplane)	Content (g/cc)	Worth Replacing Radial Blanket in Channel (lh)
-25 to +25	<u>Radial Blanket:</u> 8.56 Depleted Uranium 1.10 Steel 0.34 Aluminum 1.04 Molybdenum	0
-26 to +26	<u>Sodium Channel:</u> 1.65 Steel 0.63 Sodium	-140
-26 to +10	<u>Rod "Stem":</u> 5.9 Steel 0.14 Sodium	-15
+10 to +26	<u>Sodium Channel</u>	
-26 to +10	<u>Rod "Stem"</u>	
+10 to +24	<u>Mockup B₄C Rod:</u> 0.66 B ¹⁰ 0.19 B ¹¹ 0.36 Carbon 1.20 Steel 0.14 Sodium	-300
-26 to +7	<u>Rod "Stem"</u>	-650
+7 to +21	<u>Mockup B₄C Rod</u>	

Figure 2 shows the radial outline of the final engineering configuration in core Half No. 1 of the assembly. This loading, containing a total of $35\frac{1}{2}$ core drawers in each machine half, was just-critical with all the ZPR-III rods in. Thus, for this representation of the RAPSODIE operational loading, the critical core volume was 38.85 liters. The principal uncertainties involved in predicting the size of a RAPSODIE core of the same composition would involve the effects of fuel heterogeneity and the relative self-shielding in the B₄C rods. These effects, however, most likely would not produce a core volume in excess of a RAPSODIE design limitation of 40.5 liters.

With all of the engineering features in place (the axial blankets and gaps, and the rods and channels), another recalibration was made of ZPR-III control rods, since the aforementioned features and additional fuel loadings had changed their worths. Measurements were then made of the average

reactivity coefficients of materials throughout the core. These measurements involved two series of substitution experiments: in one series, substitutions of materials were done in a distribution of 9 drawers in each half, representing a fourth of the core drawers, excluding the center drawers (P-16); in the other series, substitutions of materials were made in the central drawers of each half. Table V presents the results of the measurements for both series and for the combined average worths in the core.

Table V. Reactivity Worths of Materials in Core of RAPSODIE Engineering Mockup

Material	Reactivity Coefficient (Ih/kg)		
	Distributed Drawers of Quadrant	Central Drawers (P-16)	Average in Core
Depleted Uranium	24	26	24
Steel	27	13	27
Sodium	110	110	110
Iron	22	10	22
Fe ₂ O ₃	31	44	31
Aluminum	67	57	67
Al ₂ O ₃	57	91	58
Oxygen	50	130	52

Worths of steel and sodium were measured in the upper and lower end gaps by substitutions of empty cans for sodium-filled cans in a distribution of drawers and then replacing the empty cans with steel. Table VI gives the results of these experiments.

Table VI. Worths of Sodium and Steel in End Gaps of RAPSODIE Mockup

Zone	Reactivity Coefficient (Ih/kg)*	
	Stainless Steel	Sodium
Lower End Gap (3 in.)	21	90
Upper End Gap (7 in.)	12	38

*Worths of materials relative to void.

A series of substitution experiments were initiated to obtain the worth of sodium in the core as a function of radial zone and axial position.

2. ZPR-VI

Measurements of the sodium versus void effects on reactivity were continued with Assembly No. 2, a 600-liter carbide system.

Reactivity changes resulting from substituting Type 304 stainless steel for voids were measured in ZPR-VI Assembly No. 2. The measurements were made as a function of radial position (measured in increments of drawers which are approximately 2 in. wide) for each of three axial positions. An axial section through the reactor is shown in Figure 3; the three positions are shown superimposed and do not show individual drawers. Figure 4 shows a plot of the radial worth and Figure 5 a representation of the axial worth. Although it is difficult to assign error limits to these measurements, because of the limited redundancy of the data, error limits of ± 0.4 lh/kg were chosen to give an indication of the possible precision; the actual errors may be somewhat less than this.

Figure 3. Axial Section through Assembly No. 2

The loss of reactivity due to table separation for an air-filled gap and a stainless steel-filled gap was measured. The results for Assembly No. 2 are shown in Figure 6. These may be compared with similar measurements performed with Assembly No. 1, previously shown in the Progress Report for November 1963, ANL-6808, p. 6.

3. ZPR-IX

a. Assembly No. 1. The availability of the ZPR-IX Assembly No. 1 provided an opportunity to isolate and study the effect of a light metal reflector. Except for the aluminum reflector (and the relatively minor core volume fraction due to the matrix), this assembly was identical with the ZPR-III Assemblies No. 11 and No. 22 and the ZPR-VI Assembly No. 1.

Figure 4
Variation in Reactivity with
Size of Gap

Figure 5. Axial Worth of Type 304 Stainless Steel

Figure 6. Radial Worth of Type 304 Stainless Steel

The first objective was to determine how accurately the reactor parameters could be predicted by using a single set of multigroup aluminum cross constants for core and reflector. The second objective was to determine the sensitivity of predicted parameters to changes in the aluminum multigroup constants. Three sets of independent calculations were carried out. These were identical in all respects (i.e., code, mesh points, convergence, criteria, etc.) except for the aluminum constants. These constants included:

- (1) The standard aluminum set (mat No. 19) from ANL library No. 201. This is a modified Hanson and Roach compilation.
- (2) The standard aluminum set (mat No. 7) from ANL library No. 135. This is a modified Yiftah-Okrent-Moldauer compilation.
- (3) An ELMOE computed aluminum (self-shielded) set (mat No. 16) from ANL library No. 135. These aluminum constants were obtained from an analysis of an assembly (ZPR-III, No. 23) containing approximately 45 v/o aluminum.

The critical mass was calculated with the RE-122 code in spherical geometry by means of the three separate sets of aluminum constants. The results for the ZPR-III Assembly No. 22 (depleted uranium reflected) and the ZPR-IX Assembly No. 1 are shown in Table VII. Assembly No. 22 served as a normalizing point and with its use led to a prediction of 283 kg U^{235} for the pre-experiment critical mass of Assembly No. 1.

Table VII. Comparison of Experimental and Calculated Critical Masses (kg U²³⁵)

	Experimental	Set No. 135					
		Set No. 201		Standard Aluminum		Self-shielded Aluminum	
		Calculated	Corrected	Calculated	Corrected	Calculated	Corrected
ZPR-III Assembly No. 22	239	280.5	240	255		255	
ZPR-IX Assembly No. 1	267	332	283	239	225	274	257

* The correction refers to a straightforward normalization of the calculated Assembly No. 1 value to the experimental Assembly No. 22 critical mass. Thus, for the Set No. 201, which quite consistently overestimates critical masses of assemblies containing U²³⁸, the correction factor is 0.852, and for Set No. 135 the correction factor is 0.94.

The experimental core edge worth was 24 lh/kg. By using a calculated core-averaged U²³⁸ fission fraction of 0.304, an experimentally determined ratio of 7 was obtained for the term $(\Delta M/M)/(\Delta k/k)$. The equivalent calculated values (using cross-section set No. 201) were 21.6 lh/kg for core edge worth and 7.4 for the ratio $(\Delta M/M)/(\Delta k/k)$.

Both critical mass and core edge worth data demonstrate the decreased effectiveness of the aluminum as compared with the depleted uranium reflector. For these assemblies, the reflector interchange (from U²³⁸ to aluminum) resulted in 1.8% $\Delta k/k$ reactivity loss.

A comparison of calculated (based on perturbation theory) and experimental values of central worth coefficients for various materials is presented in Table VIII. The table illustrates the tendency of the calculation method to underemphasize the effect of the aluminum reflector; that is, since the ZPR-IX Assembly No. 1 core is ~11% larger in volume than the ZPR-III Assembly No. 22 core, on first approximation the absolute values of the central material worth coefficients would be expected to be smaller for the ZPR-IX No. 1 assembly. As shown, this is the case for the experimentally determined values for U²³⁵, U²³⁸, and tungsten, but does not hold true for the calculated values.

Table VIII. Comparison of Calculated and Experimental Central Material Worths

Material	ZPR-IX Assembly No. 1, lh/kg		ZPR-III Assembly No. 22, lh/kg	
	Measured	Calculated	Measured	Calculated
U ²³⁵	+240	276	270	265
U ²³⁸	-10.9	-17.9	-11.7	-13.5
Tungsten (No. 37)	-22.6	-26	-24.0	-27.3
WO ₃	-24.3	-21.4	-	-
B ¹⁰	-2840	-2645	-2760	-2755

b. Assembly No. 2. The ZPR-IX Assembly No. 1 has been replaced with Assembly No. 2. The loading of Assembly No. 2 was accomplished by successively replacing a drawer containing Assembly No. 1 core

composition with the desired second core composition. The second core composition in each core drawer was composed of ten $\frac{1}{8}$ -in. columns of U^{238} , two $\frac{1}{8}$ -in. columns of U^{235} , and four $\frac{1}{8}$ -in. columns of tungsten. One octant of the core was replaced by the second core composition at a time. Table IX summarizes the loading order carried out on Assembly No. 2

Table IX. Loading Order for ZPR-IX Assembly No. 2

<u>Loading</u>	<u>Added U^{235} in Assembly No. 2, kg</u>	<u>Total U^{235} in Core, kg</u>	<u>Total Excess Reactivity, lh</u>
Assembly No. 1 (Reference)		281.15	68.53
1st Octant	14.508	295.66	73.89
2nd Octant	14.4479	310.11	88.75
3rd Octant	14.5270	324.63	110.13
4th Octant	14.8796	339.51	153.55
Central Drawer	0.2693	339.24	141.75
Two Half-drawers from Periphery	-1.6482	337.60	98.34
5th Octant	14.1546	351.75	107.93
6th Octant	13.8137	365.56	134.08
7th Octant	14.1828	379.74	158.60
8th Octant	13.8127	393.56	178.40
10-in. Long Central Drawer	-0.0062	393.55	163.71
12-in. Long Central Drawer	+0.2693	393.82	170.20
Two Half-drawers Removed from Periphery	-1.6482	392.17	137.69
Final Cylindricized Core with 1-in. Void in Center		395.6372	170.92

The near linearity of response of this loading procedure indicates the possibility of studying other core materials in a one-eighth mockup geometry and subsequent linear extrapolation to full loading conditions.

4. Instrumentation Development

a. Solid-state Neutron Spectrometer. The acquisition of two channels of suitable counting equipment and a fast coincidence unit has led to the resumption of work on the solid-state fast neutron spectrometer. Each channel consists of a charge-sensitive preamplifier, a main amplifier, and a biased post-amplifier. Pulse-shaping is either "RC" or "double delay line."

The resolution of the spectrometer is much improved over that obtained with earlier versions. For thermal neutrons, the "Q" of the $Li^6(n, \alpha)H^3$ reaction, on which this model of the spectrometer is based, has

been measured with an accuracy of 33 keV (full width at half-maximum). This is less than half of the previous uncertainty and for this experiment is about the limit set by theoretical considerations of amplifier noise.

Measurements of the lifetime of the surface-barrier diodes in a fast neutron flux have been made. Exposure of a pair of the diodes to a total of 3.4×10^{12} fast neutrons/cm² resulted in a 50% increase in width of response to thermal neutrons and a 1% decrease in average pulse height. Some preliminary efforts at measuring an in-core fast spectrum have been made. Mathematical analysis of the raw data to extract the neutron spectrum is under way.

b. Rossi-alpha. A Rossi-alpha transistorized time-analyzer design is approximately 95% complete; the construction is approximately 35% complete. This analyzer, essentially a transistorized version of the TA-15 and TA-16,² is constructed with modular units for ease of maintenance and repairability. Each electronic decade scaler in the readout has been tested and found to have a pulse-pair resolution of less than 0.33 μ sec with a power supply variation of $\pm 20\%$. All circuitry for the analyzer will be tested over this supply variation and a 50°C temperature rise to insure reliability. The modular technique developed for this analyzer has worked extremely well, and considerable use of this packaging procedure is anticipated in the future.

B General Fast Reactor Fuel Development

1. Metallic Fuels

Uranium-plutonium alloys are being investigated for use as fertile-fissile fuel materials in fast breeder reactors because many combinations of the base elements have good neutron yield 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 reprocessing 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. U-Pu and U-Pu-Fz

(1) Thermal Properties. The heat contents of U-10 w/o Pu and U-10 w/o Pu-10 w/o Fz alloys (see Figure 7) have been measured in an isothermal drop calorimeter through their fusion temperatures to about

²G. S. Brunson et al., A Survey of Prompt-neutron Lifetimes in Fast Critical Systems, ANL-6681 (Aug 1963), pp. 39-42.

1190°C. In the alpha-temperature region U-10 w/o Pu has the same heat content as uranium and a slightly lower heat content than U-10 w/o Pu-10 w/o Fz. The phases indicated in Figure 7 are for the binary alloy. The comparable parts of the fizzium alloy curves are for a more complicated structure.

Figure 7

Heat Contents of U-10 w/o Pu and U-10 w/o Pu-10 w/o Fz above 25°C

In the U-10 w/o Pu alloy the $\alpha \rightarrow \beta$, $\beta \rightarrow \gamma$, and $\gamma \rightarrow \text{liquid}$ transformations occur over a range of temperatures. The heats on transforming from α to β and from β to γ are 2.9 ± 0.3 and 4.7 ± 0.5 cal/gm, respectively. These heats were calculated for the middle of the two-phase regions and agree with those of pure uranium. However, the heat of fusion of the binary alloy was 10.2 ± 1.0 cal/gm, compared with 12.2 ± 1.0 cal/gm for the pure uranium.

The phase transformations in U-10 w/o Pu-10 w/o Fz are largely suppressed because of the rapid cooling rate obtained in the drop calorimeter. Nevertheless, the heat effects recorded indicate the transformation temperatures.

The full heat content of U-10 w/o Pu-10 w/o Fz is obtained only up to the first transformation at about 535°C, because the alloys were first fully equilibrated. The heat contents above that temperature are low by the amount of heat retained in the untransformed phases. The constructed curve in Figure 7 gives the approximate equilibrium values. Extrapolation between the solidus and liquidus of the heat-content curve from below and above gives the heat of fusion for this alloy. Midway between $900 \pm 5^\circ\text{C}$ and $1030 \pm 5^\circ\text{C}$ this value is 9.5 ± 2.2 cal/gm.

(ii) Compatibility with Potential Cladding Materials. The compatibility of U-Pu and U-Pu-Fz alloys with potential cladding is being studied to help in the selection of fuel and cladding materials for future fast reactors.

The fuel-cladding combinations being studied in the temperature range from 550 to 650°C are U-10 w/o Pu and U-20 w/o Pu-10 w/o Fz with niobium and U-10 w/o Pu-10 w/o Fz with niobium, Nb-1 w/o Zr, molybdenum, vanadium, V-10 w/o Ti, V-20 w/o Ti, Type 304 stainless steel, and Hastelloy-X. The results obtained thus far are summarized in Table X.

Table X. Compatibility of U-Pu-Fz Alloys with Potential Cladding Materials

Material, w/o	Temp, °C	Interdiffusion Bandwidth (μ)			Direction
		7 Days	17 Days	42 Days	
Nb vs.	550	3	5	6	Total band width.
U-10 Pu-10 Fz	600	7	10	15	Almost 100% into
	650	14	30	80	the clad.
Nb-1 Zr vs.	550	3	6	7	Total band width.
U-10 Pu-10 Fz	600	6	9	15	Almost 100% into
	650	12	29	73	the clad.
Nb vs.	550	6	8	12	Total band width.
U-20 Pu-10 Fz	600	10	21	35	Almost 100% into
	650	23	75	172	the clad.
Nb vs.	550	nil	nil	nil	No evidence of penetration.
U-10 Pu	600	nil	nil	nil	
	650	nil	nil	nil	
V vs.	550	-	4	5	Total band width.
U-10 Pu-10 Fz	600	-	5	6	Penetration mostly into
	650	-	-	11	the fuel.
V-10 Ti vs.	550	-	1	1	Penetration into the clad.
U-10 Pu-10 Fz	600	2	3	4	Also evidence for some
	650	3	5	8	diffusion into the fuel.
V-20 Ti vs.	550	-	-	1	Penetration into the clad.
U-10 Pu-10 Fz	600	1	2	2	Also evidence for some
	650	3	3	4	diffusion into the fuel.
Mo vs.	550	4	6	9	Total band width.
U-10 Pu-10 Fz	600	5	8	11	About 50% into the clad.
	650	11	17	24	
304 SS vs.	550	2	3	4	Penetration into the clad.
U-10 Pu-10 Fz	600	5	7	10	Also evidence for major
	650	11	16	22	diffusion into the fuel.
Hastelloy-X vs.	550	-	-	-	Penetration into the clad.
U-10 Pu-10 Fz	600	5	10	18	Also evidence for major
	650	16	29	39	diffusion into the fuel.

In Figure 8 is a log-log plot of penetration into the cladding versus time for V-10 w/o Ti and V-20 w/o Ti against U-10 w/o Pu-10 w/o Fz. The preliminary data, which also appear in the table, indicate a slope of 2

for V-10 w/o Ti versus U-10 w/o Pu-10 w/o Fz, and a slope of 4 for V-20 w/o Ti. Mathematically expressed, the penetration behavior of these combinations can be represented by

$$X^2 = kT \text{ for V-10 w/o Ti versus U-10 w/o Pu-10 w/o Fz}$$

and

$$X^4 = kT \text{ for V-20 w/o Ti versus U-10 w/o Pu-10 w/o Fz.}$$

Figure 8

Penetration of V-10 w/o Ti and V-20 w/o Ti by U-10 w/o Pu-10 w/o Fz

Extrapolation of the V-20 w/o Ti curve at 650°C to one year (a somewhat risky business with sparse, preliminary data) gives a value of 7 μ for the penetration into the cladding. A two-year extrapolation yields about 9 μ . However, these values refer only to the penetration into the cladding and not into the fuel.

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 various potential jacketing materials that have been considered, vanadium-base alloys are among the more promising, and titanium is the element favored for use as an alloying element in the vanadium. The optimum titanium content in the alloy is not yet known, and it may be anticipated that the vanadium-titanium alloy most inert to sodium is not likely

to be the composition that is easiest to fabricate. The properties and fabricability of vanadium and of vanadium-titanium alloys are therefore being investigated.

Results to date indicate that compositions with 10 to 20 w/o of 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.

(i) Fabrication. Investigation of the consolidation and fabricability of V-20 w/o Ti alloy (TV-20) has continued. Analyses of turnings from the arc-cast ingot AM 21 (see February Progress Report, ANL-6860, p. 12) were as follows:

<u>Element</u>	<u>Content</u>
Titanium	19.4 ± 0.7 w/o
Carbon	387 ppm
Oxygen	360 ppm
Nitrogen	270 ppm
Hydrogen	0.4 ppm

Electrode preparation for subsequent ingots will incorporate a slightly higher charge of titanium to compensate for the slight loss in melting and bring the final ingot composition to the target content of 20 ± 1.0 w/o.

The arc-cast ingot was cropped to remove the slight secondary pipe and machined to an 8.9-cm (3.5-in.)-diameter extrusion billet. Ultrasonic inspection revealed no internal defects. This billet was canned in Type 304 stainless steel (304 SS) and extruded at 1000°C. at a 3.6:1 reduction ratio, to break up the cast structure and refine the grain size prior to extrusion to sheet bar and tube blanks.

The extruded bar was dejacketed and the TV-20 bar machined to 4.4-cm (1.75-in.)-diameter billets for re-extrusion. Nondestructive testing again showed the bar stock to be free of defects.

One billet was jacketed in 5-cm (2-in.)-diameter 304 stainless steel (SS) and extruded at a 7.2:1 ratio at 1100°C to yield bar stock for cold rolling to 0.57-cm (0.225-in.)-diameter rods. A second billet also was canned in steel and extruded at a 13:1 ratio at 1100°C through a 0.63-cm x 2.54-cm ($\frac{1}{4}$ -in. x 1-in.) sheet bar die. The resulting sheet bar has been dejacketed and will be rolled to 1.6-mm ($\frac{1}{16}$ -in.) sheet.

Three billets were drilled to give 4.4-cm-OD x 1.6-cm-ID (1.75-in.-OD x 0.62-in.-ID) cylinders for tube blank extrusion. These billets were canned in 304 SS (OD and ID) and extruded over a tool steel mandrel

at 1100°C at reduction ratios of 13:1 and 16.2:1. The billets extruded satisfactorily and the tubing (after dejacketing) will be cold worked to small-diameter, thin-walled tubing.

The use of a two-stage extrusion process - an ingot break-down and a second product-oriented extrusion - yields a fine-grained material with very minor surface striations. This undesirable surface condition plagued single-extrusion results reported earlier.

(ii) High-temperature Mechanical Properties. In Table XI are compared the preliminary results of short-time tensile tests with V-10 w/o Ti and V-20 w/o Ti for temperatures up to 800°C with the results

Table XI. High-Temperature Mechanical Properties of Refractory Metals

Temp (°C)	Ultimate Tensile Stress (kg mm ⁻²)	Yield ^(a) Stress (kg mm ⁻²)	Elongation ^(b) (%)	Uniform ^(c) Elongation (%)	Reduction of Area (%)	Elastic Modulus (10 ³ kg mm ⁻²)
Nb-1 w/o Zr, Stress Relieved at 850°C for 1 hr; Average Strain Rate, 2.7 x 10 ⁻⁴ sec ⁻¹						
RT ^(j)	53.2	47.4	28	7.3	90	12.4
640	43.4	42.0	19	1.9	93	8.4
650 ^(d)	48.2	44.2	19	1.9	87	12.4
800	36.2	34.6	24	1.3	83	12.4
850	36.2	34.4 ^(e)	14 ^(f)	(g)	87	(g)
950	35.9	34.8 ^(e)	15 ^(f)	(g)	90	(g)
Nb-1 w/o Zr, Recrystallized at 1200°C for 1 hr; Average Strain Rate, 2.7 x 10 ⁻⁴ sec ⁻¹						
650	26.9	24.7	34	7.9	98	13.0
V-10 w/o Ti, Extruded 1110°C, Recrystallized Structure; Average Strain Rate, 2.7 x 10 ⁻⁴ sec ⁻¹						
RT	48.4	37.2	38	16.4	62	23.9
RT	50.6	39.5	38	17.3	61	14.3
650 ^(h)	49.9	45.0 ^(e)	34	15.2	53	(g)
750	37.3	27.1	42	10.7	69	8.1
V-20 w/o Ti, Extruded 1110°C, Recrystallized Structure; Average Strain Rate, 2.7 x 10 ⁻⁴ sec ⁻¹						
RT	70.3	62.6	34	16.7	56	17.7
RT	70.6	64.2	36	20.2	56	18.4
650	66.2	45.8	37	15.6	44	12.0
750	46.3	38.3	36	7.0	56	14.3
800	41.5	35.9	48	5.0	50	9.6
Mo-0.5 w/o Ti-0.08 w/o Zr, Stress Relieved at 1200°C for ½ hr; Average Strain Rate, 2.1 x 10 ⁻⁴ sec ⁻¹						
RT	77.6	77.5	(i)		(i)	
RT	79.2	63.0	2.0		1.9	
RT	79.3	75.3	19.5 ⁽ⁱ⁾		18.3 ⁽ⁱ⁾	
RT	83.7	74.2	12.5		10.5	
350	64.9	57.4	14 ^(f)		85	
350	62.4	59.9	22		88	
450	58.9	57.1	19.5 ^(f)		86	
650	56.0	49.1	23		86	
650	57.4	55.0	22		89	
650	55.7	55.0	17.7 ^(f)		85	
800	53.8	49.7	20		85	

(a) Measured at 0.002 strain offset.

(b) Measured as change in 2.54-cm gauge length after specimen is removed from furnace.

(c) Total strain at maximum load.

(d) Specimen stress relieved at 850°C for 2 hr in a different vacuum furnace.

(e) Measured with crosshead travel and may not correspond to 0.002 offset yield stress measured with extensometer.

(f) Measured as change in total length of reduced cross section - 3.18 cm for Nb-1 w/o Zr, V-10 w/o Ti, V-20 w/o Ti, 4.00 cm for Mo-0.5 w/o Ti-0.08 w/o Zr - after specimen is removed from furnace.

(g) Not measured in absence of extensometer.

(h) Specimen surface oxidized during test due to air leak in vacuum furnace.

(i) Test terminated before specimen failure.

(j) RT - room temperature.

of similar tests with Nb-1 w/o Zr and Mo-0.5 w/o Ti-0.08 w/o Zr (TZM). All tests were conducted in a vacuum (2×10^{-5} mm Hg or less) at a constant crosshead rate of 8.3×10^{-4} cm/sec.

The mechanical extensometer used to measure specimen elongation is not giving satisfactory data for determination of the elastic modulus. The source of the apparent errors is being sought.

The uniform elongations of V-10 w/o Ti and V-20 w/o Ti were significantly greater than that for Nb-1 w/o Zr. The V-Ti alloys appear to get much of their strength from strain hardening, which leads to greater uniform elongation; Nb-1 w/o Zr is a dispersion-hardened alloy. The difference in behavior is also evidenced by the difference between the yield and ultimate tensile stresses, as well as by the strain hardening exponent which, at 650°C, is 0.028 for recrystallized Nb-1 w/o Zr and 0.14 for V-20 w/o Ti. At temperatures above 650°C, V-10 w/o Ti and V-20 w/o Ti show a sharp decrease in both uniform strain and tensile strength. Apparently at the higher temperatures the ability to strain harden decreases.

(iii) Corrosion by Sodium. Corrosion data for vanadium samples exposed to flowing sodium at 650°C for 9 days were reported in Progress Report for February 1964, ANL-6860, p. 4. The oxygen concentration in the sodium was ~200 ppm, not ~50 ppm as reported. The samples were in two conditions of heat treatment and contained two different initial oxygen concentrations. The results of the bend tests, consisting of loading the specimens in compression until fracture occurred, are summarized in Table XII.

Table XII. Effect of Exposure of Vanadium at 650°C to Sodium Containing Approximately 200 ppm Oxygen on Room-temperature Bend Ductility

Sample*	Included Angle at Fracture**	Sample*	Included Angle at Fracture**
Stress Relieved, 1070	153°	Stress Relieved, 200	150°
Recrystallized, 1070	140°	Recrystallized, 200	151°

*The number refers to initial oxygen concentration, ppm.

**Duplicate samples not exposed to sodium survived a complete bend (included angle of 0°) without fracture.

All the samples exposed to the sodium failed in a brittle manner. Microhardness measurements indicated that oxygen had penetrated the entire sample thickness, about 0.08 mm.

Microhardness measurements under reduced load indicate that the V-20 Ti alloys have a contaminated layer about $15\ \mu$ thick, as compared with a $100\text{-}\mu$ layer produced on the V-10 Ti alloys tested under identical conditions.

The calculations for metal reacted reported in ANL-6860 were based, among other things, on the assumption that all oxygen goes to the formation of a VO film. This assumption leads to a film thickness after a 9-day exposure of about $2.5\ \mu$. In fact, metallographic examination reveals a "disturbed" layer about $25\ \mu$ thick (corresponding to hardness increase of >100 DPH). This indicates there is little, if any, VO formation; rather, this indicates solution of oxygen in a narrow region.

Equivalent "rates of metal loss" for V-Ti alloys that lost weight upon exposure to sodium were reported in ANL-6840, p. 27. These values were in error and corrected values are given in Table XIII.

Table XIII. Results of Exposure of Vanadium Alloys to Flowing Sodium at 650°C and 195 ppm Oxygen

Alloy, w/o	Rate of Wt. Loss, mg/cm ² /mo	Equivalent Metal Loss	
		cm/mo $\times 10^3$	mils/mo
V-10 Ti (SR)*	3.23	0.92	0.37
V-10 Ti (RX)**	2.76	0.76	0.29
V-10 Ti (SR)	3.33	0.72	0.29
V-20 Ti (RX)	3.72	0.83	0.32

*Stress Relieved.

**Recrystallized.

At present, there is no explanation for the occurrence of weight losses in one test and weight gains in a second test (under very similar conditions) of vanadium and vanadium-titanium alloys. The appearances of the samples indicate that weight gains would be expected, and therefore experimental errors are suspected in the first test.

(iv) Compatibility with Molten Fuels. Safety considerations for EBR-II and other future fast reactors require information about the rate of penetration of prospective cladding materials by molten uranium and plutonium-containing fuels.

Preliminary results about the compatibility of vanadium with molten uranium, U-10 w/o Pu, and U-10 w/o Pu-10 w/o Fz in the temperature range from 1100 to 1400°C are plotted in Figure 9. The effect of uranium and U-10 w/o Pu on vanadium differs from that of molten U-10 w/o Pu-10 w/o Fz. The penetration rates of uranium and U-10 w/o Pu vary

linearly with temperature, being slightly greater for U-10 w/o Pu. The fissionium-containing alloy does not behave as predictably. Their behavior may, however, be explained by studying the various binary phase diagrams between vanadium and the melt components. Compounds are present in the Ru-V and the Zr-V systems. Experience with the uranium-iron system has shown that compound formation during penetration can have a profound effect on the penetration rate. The penetration rates in vanadium versus U-10 w/o Pu-10 w/o Fz are much slower ($2 \mu/\text{sec}$ at 1150°C) than those found in Type 304 stainless steel versus U-5 w/o Fs ($300 \mu/\text{sec}$ at 1150°C). In vanadium versus U-10 w/o Pu-10 w/o Fz it seems likely that the presence of Zr-V and Ru-V compounds are responsible for the rate reversal at 1200°C and for the sudden increase in rate at about 1300°C , which happens to correspond to the 1300°C melting point of ZrV_2 .

Figure 9. Penetration of Vanadium by Molten Uranium, U-10 w/o Pu and U-10 w/o Pu-10 w/o Fz

(v) Nondestructive Tests. Several specimens of V-20 w/o Ti alloy (TV-20) were examined ultrasonically, and no defects were detected. The specimens included a casting 8.97 cm in diameter, two solid cylindrical extrusions 4.45 cm in diameter, and five tubular extrusions 4.42 cm in outer diameter and 1.60 cm in inner diameter.

b. Niobium Alloys

(i) High-temperature Mechanical Properties. The mechanical properties of Nb-1 w/o Zr alloy at room temperature to 800°C are compared with those of TV-10, TV-20, and TZM in Table XI.

(ii) Nondestructive Tests. Thirty tubes of Nb-1 w/o Zr alloy, totalling 30.48 m in length, were found by ultrasonic longitudinal tests to have many laps on both the inner and outer surfaces. These tubes were 4.5 mm in outer diameter and 0.5 mm in wall thickness.

c. Molybdenum Alloys. The mechanical properties of TZM at room temperature to 800°C are compared with those of niobium-base and vanadium-base alloys in Table XI.

d. Tungsten-coated Stainless Steel Tubing. Radiographic examination of 2.44 m of tungsten-coated stainless steel tubing revealed defects that were cause for rejection. This tubing had an inner diameter of 4.22 mm and a wall thickness of 0.23 mm.

3. Irradiation of Experimental and Prototype Fuels

a. Development and Fabrication of Experimental EBR-II-type Irradiation Specimens. A total of 24 U-10 w/o Pu-10 w/o Fz (93.2 w/o U²³⁵) alloy rods were injection-cast into Y₂O₃-coated Vycor molds. The rods appear to be of good quality and are being radiographed before machining and fuel-rod assembling.

A total of 53 (28 additional during the reporting period) of 55 scheduled fuel rods were assembled and bonded. Twenty-four of these have been inspected for sodium level and bond quality and seventeen were found to be acceptable. Difficulties experienced in the interpretation of results of the eddy-current bond test for the remaining rods are being resolved with the use of additional new test standards. Rods having a sodium level outside the specified range but showing an acceptable sodium bond are being accepted.

Processing procedures and equipment are being prepared for the loading of experimental fuel rods into 100-cm secondary capsules.

4. Powder-loaded Radiator-type Fuel Elements

The term "radiator fuel element" is a name given to an extended-surface heat-exchanger fuel concept. Theoretically, these can function at higher powers for a given fuel temperature than plate- or rod-type fuel elements. The complexity of solid shapes required to fill such elements has delayed the development of the radiator fuel elements. These studies are directed toward an understanding of the compacting of powdered material and providing specimens for irradiation of powdered elements preparatory to fabricating vibratory compacted radiator shapes.

a. Vibratory Compaction. Data for the vibratory compaction of irregularly shaped particles of one size classification were submitted to the Applied Mathematics Division (AMD) for curve fitting. Before releasing

the work to the AMD, it was found that the packing efficiency of irregular shapes is proportional to the particle size. The function holds for particle "diameters" down to 0.25 mm and is of the form $Pe = A + (K/d_1)$, where Pe is the packing efficiency, A is the limiting density, d_1 is the "diameter" of the single size classification, and K is a constant. The above fraction was found for the region where the boundary condition is negligible. The boundary condition has been found to be expressed by the function Be^{-CD/d_1} , where B and C are constants, and D/d_1 is the ratio of diameter of container (D) to "diameter" of the single component (d_1). AMD will find the values of A , K , B , and C by least squares in an equation of the form $Pe = A + (K/d_1) + Be^{-CD/d_1}$.

A plot of Pe versus d_1/d_2 in the binary system of irregular shapes was found to be of the same shape as that for a spherical system. AMD will fit the data to a least-squares equation of the form $Pe = A + Be^{-Cd_1/d_2}$, where d_1/d_2 is the ratio of the first component "diameter" (d_1) to the "diameter" of the second component (d_2).

Work has been started on the packing of ternary systems of irregular shapes.

5. Zero Power Reactor Fuels

a. Fuel Experiments. Very large numbers of small fuel elements are required for zero power reactor experiments. These must be repeatedly loaded and unloaded into various test configurations to determine the effects on reactivity and other physical characteristics. The durability and fire resistance of the elements is important. With plutonium-containing fuel, the possibility of fuel-element rupture is a serious health hazard. A plutonium fuel element fire would be disastrous. The following experiments were performed to provide fuel elements that can be evaluated for durability and fire resistance.

(i) U-18 w/o Pu-3.8 w/o Fe. Three precision casts were made into ZPR-III-size plate molds. A melt temperature of 1325°C was maintained, and mold temperatures of 100°C, 300°C and 550°C were used. The intermetallic compound appeared to be brittle and prone to cracking. Castings did not shrink away from the mold surfaces upon cooling and tended to warp.

(ii) U-18 w/o Pu-1.5 w/o Fe and U-18 w/o Pu-2.5 w/o Mo. Rods, 1 cm in diameter and 16 cm long, were gravity cast into a Y_2O_3 -coated split copper mold. A mold temperature of 300°C was used. Cast surfaces were good and a fine grain size was apparent in fractured castings.

b. Properties of Zero Power Fuels. Chill-cast specimens of U-27 a/o Pu alloys containing 6 a/o of various additional alloying elements

(see Progress Report for September 1963, ANL-6784, p. 13, and Progress Report for November 1963, ANL-6808, pp. 70 to 71) have been exposed for 4 mo to flowing room air at 15 cfm, 24°C, and 25 to 56% relative humidity. Table XIV summarizes the test results, and Figure 10 shows the appearance of the specimens at the end of the test.

Table XIV. Results of Corrosion in Air of Uranium and U-Pu Base Alloys

Alloy (a/o)	Original Weight (gm)	% Weight Gain after				Remarks
		1 mo	2 mo	3 mo	4 mo	
U-27 Pu-6 Al	6.37	13.2	15.1	15.9	16.0	Began gaining weight on 2nd day; completely disintegrated by 6th day.
U-27 Pu-6 C	3.23	12.1	14.8	15.5	15.8	No gain in weight first 5 days. Gained 0.03 gm by 6th day. Completely disintegrated by 6th day.
U-30 Pu	6.85	9.5	13.0	15.2	15.8	Began gaining weight on 2nd day. Completely disintegrated by 6th day.
U-27 Pu-6 Cu	4.45	7.6	11.7	14.8	15.7	Showed some signs of swelling and initial disintegration on 6th day. Began gaining weight on 7th day. Completely disintegrated by 21st day.
U-27 Pu-6 Fe	2.95	0	0	0.7	1.0	Began to disintegrate after 11 weeks, but did not start gaining weight until after 12th week. Some powder after 4 mo.
U-27 Pu-6 Zr	3.75	0	0	1.3	2.1	Began gaining weight shortly after 2 mo, but did not start disintegrating until after 3 mo. More powder than Fe alloy.
U-27 Pu-6 Mo	3.49	0	0	0	0	A very thin surface film spalled off after 3 mo, but no weight change.
U-20 Pu-10 Fz*	0.10	0	0	0	0	No weight change. Surface tarnished.
U	3.19	0	0	0	0	No weight change. Surface tarnished.

*U-20 Pu-10 Fz in w/o.

The U-30 a/o Pu alloy and the ternary alloys containing aluminum and carbon completely disintegrated to a powder in six days. The copper alloy started to powder in six days and was all powder in 21 days. The Fe, Mo, and Zr alloys (as well as the U and U-20 w/o Pu-10 w/o Fz) were slightly tarnished but showed no appreciable weight change after 30 days. At the end of 4 mo the molybdenum alloy had not changed weight and was only slightly corroded, whereas the iron alloy, and to a greater extent the zirconium alloy, showed appreciable powdering.

Figure 10. Appearance of Specimens after 4-mo Exposure in Air. Top row: U-27 a/o Pu-6 a/o Al, U-27 a/o Pu-6 a/o Fe, U-27 a/o Pu-6 a/o Zr, U-27 a/o Pu-6 a/o Cu. Extreme right: Uranium. Bottom row: U-27 a/o Pu-6 a/o Mo, U-27 a/o Pu-6 a/o C, U-30 a/o Pu, U-20 w/o Pu-10 w/o Fz.

Ignition studies (by the ANL Chemical Engineering Division) of specimens from the above chill castings showed a similar trend - the molybdenum and iron alloys had high self-heating temperatures and the others were relatively low.³ A more recent test of the chill-cast iron alloy after several months of storage resulted in a much lower ignition temperature, suggesting that this alloy may deteriorate with time, making it an undesirable zero power fuel.

Of the more promising alloying elements, iron was considered the least undesirable because it will be present in the structural members of fast reactors. Therefore iron alloys were studied further. The above specimens had been chill cast to give very fine grain structures. Much coarser-grained, slower-cooled castings of the iron alloy were also ignition tested and found to have relatively low ignition temperatures. Metallographic and electron microprobe analysis showed that the coarse-grained specimens consist of two phases (about 50% each). One phase contained 73 w/o U, 24 w/o Pu, and 3 w/o Fe, and appeared to be based on the $(U,Pu)_6Fe$ compound. The other contained no iron and is the zeta phase with 36 to 38 w/o Pu, balance uranium.

We are studying further the influence of composition and casting method in an effort to find the best set of conditions for making U-Pu zero power metal fuels.

³Progress Report for November 1963, ANL-6808, p. 70.

The linear thermal expansion of the U-27 w/o Pu-6 w/o Fe alloy (injection cast) was studied through four thermal cycles from room temperature to 632°C in a quartz pushrod dilatometer. The four runs gave very similar expansion curves, from which average linear expansion coefficients were as follows:

Temperature Range (°C)	Linear Expansion Coef/°C
20-375	8.7×10^{-6}
375-565	16.1×10^{-6}
20-600	12.9×10^{-6}

A sharp transformation occurred at 573°C on heating and 541°C on cooling.

C. General Fast Reactor Fuel Reprocessing Development

1. Melt Refining of Uranium-Plutonium-Fissium Fuels

The use of chloride flux in the melt refining process has given higher yields and allowed lower operating temperatures in small-scale experiments than has melt refining in the absence of flux. In the absence of flux, spent EBR-II fuel material is maintained at 1400°C in a zirconia crucible for 3 to 4 hr, and the purified metal is poured into a mold; approximately two-thirds of the fission products are removed during this treatment through volatilization and selective oxidation. Process conditions for chloride flux-melt refining are currently being evaluated.

In the chloride flux-melt refining procedure, the fuel material is melted beneath a chloride flux in a dense, high-purity beryllia crucible. The fuel charge in the present tests is about 700 g of 81 w/o U-10 w/o Pu-9 w/o fissium alloy. Currently, about 200 g of an 81 w/o BaCl_2 -19 w/o CaCl_2 flux containing magnesium chloride as a rare earth oxidant is used with each fuel charge. Cerium is alloyed with the metal at a concentration of about 0.6 w/o as a stand-in for rare earth elements. The materials are heated and are maintained at 1150°C for 1 hr under argon. The rare earths in the fissium are preferentially oxidized into the salt phase in this step. The metal phase freezes as the materials are allowed to cool to 860°C, at which temperature the still-molten flux is poured off. The purified metal is then heated to 1300°C and poured into a mold.

In three preliminary runs, the factor for separation of rare earths from plutonium (defined as the ratio of cerium to plutonium in the final flux phase to the same ratio in the final metal phase) ranged between 4100 and 7400 as follows:

Excess Oxidant (%)	Cerium Removal from Ingot (%)	Plutonium Transfer to Flux (%)	Plutonium Vaporized (%)	Separation Factor
10	98.2	0.45	~0.002	7400
10	97.6	0.29	~0.006	7200
100	99.1	1.88	~0.088	4100

The high retentions of plutonium in the metal phase, particularly where a 10% excess of oxidant was employed, is encouraging. Work is continuing to reduce further the transfer of plutonium to the flux phase.

2. Skull Reclamation Process

The skull reclamation process will be used for the recovery and purification of fissionable material that remains in the melt refining crucible after a melt refining operation. The steps include: noble metal extraction, uranium reduction, precipitation of a uranium-zinc intermetallic compound, intermetallic decomposition, uranium dissolution, and (after removal from the primary vessel to a separate vessel) retorting of the uranium product solution to concentrate the uranium. Six skull reclamation demonstration runs have been performed through the uranium dissolution step in large-scale (2.5 kg of skull oxide) integrated equipment. Analyses of samples from waste streams are not yet complete for all runs.

Samples of waste ingots from three early demonstration runs (see Progress Report for September 1963, ANL-6784, p. 23) have been analyzed for uranium and fission product elements of interest. The analytical data obtained for the waste ingots, along with analytical data for the waste flux phase, have been used in determining the effects of agitation on reaction rates and fission product removal. In the first step of the process, noble metals are extracted by zinc from skull oxide suspended in a halide flux, and then the zinc phase is removed as waste. Molybdenum and ruthenium are present in the zinc phase in amounts which greatly exceed their solubility at the transfer temperature. Results showed that when the zinc solution was transferred without agitation or at a low agitation rate, removal of molybdenum and ruthenium from the system was low (21 to 59%). Preliminary results for subsequent runs indicate that, with an agitation rate which keeps the molybdenum and ruthenium in suspension during transfer, considerably higher removals (approximately 90%) of these elements are achieved.

Following noble metal extraction, uranium oxides suspended in molten halide salt are reduced to metal by 5% magnesium-zinc solution. Results for this step indicate that at an agitator speed of 900 rpm, $\frac{1}{2}$ hr was required for complete reduction of uranium oxide, whereas at 500 rpm, 2 hr were required.

Uranium material balances for these three runs were reasonably good (97.5 to 108%), and 91 to 103% of the uranium was accounted for in the final uranium product solution.

3. Advanced Processes

The substitution of a ternary cadmium-zinc-magnesium alloy for magnesium-zinc as a medium for the processing of uranium-plutonium-fizzium fast reactor fuels may permit the use of lower temperatures and stainless steel equipment. The distribution coefficients for praseodymium between $MgCl_2$ -30 m/o $NaCl$ -20 m/o KCl and Cd - Mg - Zn alloys have been determined as a function of the composition of the metal phase at 600°C (see Figure 11). Praseodymium was chosen for study because it has been found to be the rare earth that is most difficult to separate from plutonium (see Progress Report for April 1962, ANL-6565, p. 27). During most of the measurements of the distribution coefficients, the liquid metal was kept saturated with uranium. Minimum values of the praseodymium distribution coefficient are about 0.06 at 20 a/o magnesium in the Zn - Mg system and about 1.3 at 38 a/o magnesium in the Cd - Mg system. These results, combined with data obtained from earlier work, indicate that separation of plutonium from rare earths by means of a molten salt-molten metal system in stainless steel equipment is feasible.

Figure 11
Distribution of Praseodymium between $NaCl$ - KCl - $MgCl_2$ Molten Salt Alloy and Cadmium-Zinc-Magnesium Alloy

Conditions	
Temp:	600°C
Crucible:	Tantalum
Atmosphere:	Argon
Flux:	30 m/o $NaCl$ -20 m/o KCl - 50 m/o $MgCl_2$

The solubility of plutonium as a function of temperature in molten alloys of the nominal compositions: Cd-20 a/o Zn, Cd-20 a/o Zn-10 a/o Mg, Cd-20 a/o Zn-15 a/o Mg, and Cd-20 a/o Zn-20 a/o Mg, has been determined. The solubility increased with increasing zinc concentration and with increasing magnesium concentration in the cadmium-rich region of the ternary solvent. In general, plutonium has a much higher solubility in the alloys studied than does uranium. For example, at 600°C in Cd-20 a/o Zn-15 a/o Mg, the solubility of uranium is about 3 w/o and that of plutonium is about 13 w/o.

A program is underway to develop engineering technology applicable to the separation of plutonium from rare earth fission products by multi-stage extractions in molten metal-salt systems. A prototype high-temperature (~600°C) extraction column is being designed that will be used to determine extraction rates and column behavior for the systems studied.

A 1-in.-ID, 21-in.-long glass column is being operated at low temperatures to obtain preliminary qualitative information applicable to the high-temperature extraction column. In two runs, Wood's metal (a Cd-Bi alloy: mp, 70°C; density, 9.4 g/cm³) was made to flow countercurrently to water through packing consisting of either 3.5-mm dia, 4-mm long half-cylinders of nickel or 5-mm glass spheres. It was observed that metal hold-up in the column was high. Metal was held in the form of pools which grew to a critical size and then discharged rapidly to pools lower in the column. The transfer of solute elements between phases will very likely depend upon the pattern of metal movement and the size of the pools formed. In order to obtain some quantitative information on mass transfer rates between salt and metal phases, an experiment is being performed in which cadmium-magnesium is passed through a static column of chloride salt which contains uranium chloride. The metal will be collected and analyzed for uranium.

4. Materials and Equipment Evaluations

Tungsten is the material of construction of the crucible presently used to contain zinc-magnesium-flux systems of the EBR-II skull reclamation process. Crucibles now in use have been fabricated by pressing and sintering of tungsten powder. A 4-in.-OD, 6-in.-high crucible which was fabricated* by spinning from a tungsten sheet is being tested under conditions of the skull reclamation process. No observable changes were noted as a result of heating a zinc-flux charge to 800°C in the crucible, maintaining that temperature for 4 hr, cooling to 525°C (below the solidification temperature of the flux), reheating to 800°C, and pouring.

*Fabricated by Metalwerk Plansee, Reutte, Tirol, Austria.

A beryllia or beryllia-lined crucible is required for the retorting step of the skull reclamation process. Two silicon carbide (Type KT and Crystolon 63) rings which had been coated with beryllia by plasma spraying were subjected to 6 thermocycles between 220°C and 900°C under argon. They appeared to be unaffected by the thermal cycling.

D. EBR-II

1. Reactor Plant

The primary-system bulk sodium was heated electrically from 270°F to 580°F over a five-day period to check out the fuel-handling equipment, the primary sodium pumps, and other primary-system and auxiliary systems equipment.

a. Fuel Handling. Four verification and two filter subassemblies were loaded into the reactor. The verification subassemblies are specially cleaned, natural uranium core-type subassemblies which will be removed from the reactor after operation of the primary sodium pump. After vacuum distillation, they will be shipped to Argonne, Illinois, for disassembly and inspection.

The control rod in position No. 8 of the reactor was transferred to position No. 7 to make room for the oscillator thimble and rod assembly, which was installed. The oscillator gripper and holddown tube for the thimble were lower and attached to the rod and thimble to secure them during pump operation.

Four dummy steel-core subassemblies were removed from the storage basket, cleaned, and stored.

b. Primary Sodium Pump. Following the insertion of pump No. 2 into the primary tank, both pumps were operated for several days for initial mechanical and electrical testing. Pump speeds up to 840 rpm have been achieved with both pumps operating simultaneously. At this speed, total flow through the core and inner blanket only was about 7600 gpm. The mechanical operation of the pumps was satisfactory, and minor modifications to the electronic control circuits were undertaken.

c. Fuel Gripper. The fuel gripper and holddown were positioned over the fixed subassembly in the outer blanket in a preliminary checkout; operation was satisfactory. During fuel-handling operations, the fuel gripper operated normally, indicating that the binding problem has been corrected.

d. Control Rod Drives. The gripper jaw limit switches on several drives were adjusted for better performance.

e. Safety Rod Drives. Level alarm float assemblies were installed and placed on the safety rod drive shock absorbers. The units were filled with oil and checked out.

f. Seal Heater Control System. New blade thermocouples were installed in the large rotating plug. Half of these thermocouples control heat input during the molten condition. Checkout and adjustment of the system continued during the month.

g. Sodium-purification System. A small leak in a new weld of the plugging loop was repaired. During subsequent operation, the plugging temperature was determined to be less than 250°F. Difficulties encountered in establishing flow through the cold trap have been traced to a high-resistance connection in the dc bus supplying power to the electromagnetic pump.

h. Cover-lifting Device. A clutch was installed in the cover-lifting drive to limit the amount of torque applied to raise and lower the reactor vessel cover. The "out-of-step" circuit for the four drive screws was readjusted.

i. Fuel-unloading Machine. Minor modifications were undertaken to the fuel-unloading machine to improve its performance. A drip pan has been installed in the rotating plug to catch sodium from a subassembly or the gripper. A new stop for the rotating plug was installed to facilitate removal of the drip pan. The fuel-unloading machine coffin supports were modified to make the coffin "self-aligning" with respect to the fuel transfer port or interbuilding coffin. A new heater was installed on the fuel unloading machine to preheat the inlet argon to a higher temperature. This will permit faster preheating of a subassembly prior to lowering it into the primary tank.

A new vapor trap was installed in the argon system of the fuel-unloading machine, replacing the old trap, which had been partially plugged with sodium and sodium oxide.

j. Interbuilding Coffin. The interbuilding coffin was satisfactorily used to load the four verification subassemblies into the reactor via the fuel-unloading machine.

k. Fuel-element-rupture-detection Loop (Delayed-neutron Monitor). Welding and radiography of piping in the loop was completed. Lead shielding was fitted adjacent to the piping. The neutron detectors were installed inside graphite ingots which have been placed in their support frames.

l. Reactor Building Containment. The inflatable seal for the outer door of the personnel air lock was replaced and the lock successfully leak-tested. The hydraulic system for this air lock was modified to provide more dependable service.

The high leak rate in the purge exhaust isolation valve was found to be due to a worn rubber seat. This seat will be replaced.

2. Sodium Boiler Plant

New transition pieces were welded into the 12-in. yard lines early in the month (see Progress Report for February 1964, ANL-6860, pp. 24-5). The four circumferential field welds and the two original longitudinal seam welds (welded pipe) were inspected radiographically and found to be acceptable. Two defective welds in the 2-in. vent lines were satisfactorily reworked.

Welding, radiography, and helium leak-testing of the secondary cold trap piping and the connecting piping for the differential pressure cell of the main secondary flowmeter were completed satisfactorily.

For leak-testing of the yard piping and the differential pressure cell piping, helium, in excess of 10 v/o, was added to the argon blanket gas in the secondary system. Pressure within the system has been held at 6 in. of water.

To attain the required leak-testing sensitivity, vacuum jackets were mounted on the 12-in. pipe, enclosing the weld to be tested. The jackets were evacuated by a mechanical vacuum pump and the leak detector was connected to this evacuated line. All welds tested were acceptable.

Cladding of the yard piping with carbon steel for induction heating has been completed. The installation of insulation and induction heating wiring was nearly completed.

Resistance heaters were installed and insulation was completed for the secondary cold trap piping and for the differential pressure cell of the secondary flowmeter.

The NaK-to-air heat exchanger for cooling the temporary cold trap was replaced after removal to make working room for replacement of the permanent cold trap.

The two safety valves on the steam drum were adjusted cold for lifting pressures of approximately 1350 and 1390 psig, respectively. Previously, during hot testing with steam, they had both lifted at 1335 psig. Actual lifting pressures will be determined during the next hot operation of the steam generator.

Insulation repair required after converting the sodium-recirculating pumps to water cooling was completed.

The temperature of the secondary storage tank remained at 350°F during the month.

3. Power Plant

All power plant systems remained in shutdown status. The steam generator was maintained in wet layup as previously reported.

Installation of the turbine lube oil purification system was completed and placed in service. The purifier operated continuously for about one week with the turbine on turning gear. The oil now appears to be free of contaminants, and the purification system and lube oil transfer system are operational.

Installation of connecting piping between the feedwater and blowdown lines in the Sodium Boiler Building has been started. This line will be used for feedwater recirculation to prevent cold feedwater flow to the steam drum during startup of this system.

4. Training

Several foremen spent two weeks on approach-to-power training.

A group of technicians participated in reactor systems training, plant orientation, power plant, and coolant systems training.

Progress was made in the application of programmatic systems training for technicians on shift. This effort is important to attain and maintain proficiency at a point in time when all operations personnel are on a rotating shift basis and not available for formal daytime training.

5. Fuel Cycle Facility

During the period that the Argon Cell contained an air atmosphere, (January 31 to February 27, 1964) the removal of mercury which had been blown out of the ceiling blister seal and had fallen into the cell (see Progress Report for August 1963, ANL-6780, p. 18) was completed. About 97% of the original quantity of mercury added to the blister seal was recovered in the cell cleanup operation.

Also, during this period, mechanical and electrical components and systems of cell equipment were examined, instruments were tested, and corrective work on cell equipment was performed. These activities pertained to the following:

a. Argon-purification system. (i) All bearings on the compressor and drive motor were considered to be in satisfactory condition, (ii) the

mercury-poisoned palladium catalyst was replaced with fresh catalyst, (iii) analytical instrumentation was checked, and (iv) the entire purification system was checked for leaks and was found to be leak tight.

b. Cranes and manipulators. (i) Slip clutches were installed on the hoist motors of the three cranes, and (ii) new bridge drive shafts of improved design for the manipulators were installed and existing clutch halves were machined to fit these new shafts.

c. Melt refining furnace ("A" furnace). (i) Minor components in the furnace subassembly were replaced with components of improved design, and (ii) the 50% lead-50% tin solder (mp, 212°C) was removed from the seal trough and replaced with a higher-melting-point (260°C) solder. 88% lead-12% antimony.

d. Cell lighting luminaires. The luminaires in the Air and Argon Cells were modified slightly to assure easier remote removal and replacement of the end plate (which carries the lamp and all electrical connections) of the luminaire shell. (The 72 one thousand-watt mercury vapor lamp bulbs in the Argon Cell have been in operation for about 8000 hr; during this time, only one lamp has burned out. A measurement of the illumination in the Argon Cell showed it to be 125 foot candles, which is about 70% of the value when the lamps were new.)

On February 27, 1964, 118,000 cu ft of argon was admitted to the Argon Cell. The air concentration was reduced to about 3.3%. After the argon fill was completed, the pressure control system for the cell was placed in operation, and the cell pressure was maintained at a plus pressure of 0.5 in. of water. The argon-purification system was also started. During the day, the purification system was operated with the addition of slightly less than the stoichiometric requirement of hydrogen; during the night and over the weekend, the rate of hydrogen feed to the purification system was very low. By March 5, 1964, the concentrations of oxygen and water in the argon atmosphere were 10 and 60 ppm, respectively. The air in-leakage rate of the Argon Cell was determined in a 22-hr test, during which the cell pressure was reduced to a negative pressure of 5 in. of water. Under these conditions, the air in-leakage rate was 0.02 scfm. Since an in-leakage rate of 0.005 cfm had been previously attained (see Progress Report for September 1963, ANL-6784, p. 21), an effort is being made to detect leaks so that the present rate of in-leakage might be reduced.

Glass cover plates ($\frac{1}{4}$ -in. thick) have been installed to protect the innermost slabs (A slabs) of the viewing windows of the Argon Cell. Twelve cover plates were installed prior to the admission of argon into the cell (see Progress Report for August 1963, ANL-6780, p. 16). The remaining ten cover plates were installed after the argon atmosphere had been replaced with an air atmosphere.

The EBR-II fuel cycle requires the remotely controlled refabrication of irradiated fuel and reassembly of the fuel elements. Equipment to cast, jacket, and sodium-bond fuel rods, and to reassemble fuel elements has been constructed for this facility. The equipment is being installed and tested under this program.

All the fuel-rod fabricating equipment in the Argon Cell is operational. This includes the melt-refining equipment, the injection-casting equipment, the pin-processing equipment, and the fuel-rod settling and welding equipment. The fuel-rod leak detector installed in the air cell is also operational. The fuel-alloy ingots produced during Core-I fabrication and the hardware for the fuel-pin jackets have been received at Idaho. Fuel rods can now be made in the Argon Cell.

Difficulty has been experienced in controlling and predicting the sodium level in completed rods. A batch of twenty dummy rods was made remotely in the argon cell to verify the method of level prediction. They were leak tested and bonded in the Air Cell. X-radiography, used to study the sodium levels, showed that the rods contained bubbles in the sodium between the fuel pin and the restrainer; many contained sodium adhering to the jacket tube above the sodium level. Under these conditions, the sodium level has no significance and the accuracy of the prediction method cannot be verified. Work is now under way to determine the impact necessary to lower trapped sodium.

In-cell and subcell installations for the slag-oxidation furnace are essentially complete except for subcell leak-testing and installation of the oxygen-supply system. Minor modifications, made for ease of handling, have been completed on the slag oxide dumper, which is ready for installation in the Argon Cell.

Testing of the drive and cutter portions of the decanner in the glove-box are being made to increase the life of a decanner tool, which at present is in the order of 100-150 pins per tool, and to examine the effects of various parameters on the action of the bonding sodium. Initial results indicate that neither variations of oxygen or water content in the argon atmosphere between 10 and 400 ppm, nor heating of the scrap separator tube, reduce the power requirements or increase the slippage of the decanner unit. Tests at normal (3 in./min) forward speed and at half-speed on the decanner have as yet failed to show any improvement of tool wear or any difference between the two speeds.

A table for the decanned fuel pin samples has been built, and it and the sampler are ready for installation in the Argon Cell.

Remaining portions of the assembly machine were received during the month. The base frame of the machine was moved into the cell, and the welding assembly and force bridge drive have been installed.

Four uranium-recovery runs were made with glass scrap from the demolder station of the pin-processing machine to evaluate the feasibility and efficiency of a proposed recovery operation. The uranium scrap at the demolder station comes from two sources. The first is lengths of cast pins that are shorter than 13 in. and are rejected automatically into the scrap container at the demolding operation. The second is flash metal that adheres to the outside or the end of a glass mold. All of this material - glass, flash metal, and short castings - is diverted into one scrap container. It would be possible to pick out the larger sections of cast metal with a Model A manipulator; and, for this reason, an arbitrarily selected length of 1 in. or less has been chosen as presently unseparable. The proposed separation operations have consisted of ball-milling the mixed scrap and passing the ground product through a 170-mesh screen.

The results of four runs made on scrap from four injection casting runs are presented in Table XV. Averaging the data from the four runs shows that the total uranium scrap is about 226 g/120-mold batch and that the unseparable scrap (1 in. or less) is about 154 g/120-mold batch. The efficiency of recovery of the small scrap by ball-milling and screening is about 93-95%. A silica analysis of the +170 material from the fourth run showed a content of 400-500 mg SiO_2 for the 136 g of uranium (~3500 ppm). Although this is considerably above the present alloy specification, small amounts of such material might be blended in with low-silica alloy.

Table XV. Uranium Scrap Recovery from Crushed Vycor Molds

Run No.	Weight of Metal and Glass (g)	Weight of Uranium ^a (g)	Weight of Separable Uranium ^b (g)	Weight of Uranium in +170 Mesh (g)	% Separated	Number of Castings
1	2437	101	101	100.44	99.0	178 ^c
2	828	90.4	~60	89.0	95.6	57
3	1660	302	150	298.8	99.5 ^d	115
4	779	141.4	111	136	96.2	50

^aSum of metal on +170 mesh screen plus metal in -170 grindings (chemically determined).

^bAll scrap 1 in. or less in length.

^cCastings from two runs.

^dThis batch contained several large pieces of casting which accounts for the high separability weight.

It appears that the amount of unseparable scrap is large enough to warrant a special operation and that the ball-milling and sieving approach is adequate for this purpose.

The initial calibration runs with the pin-processing equipment have been completed. The data of Table XVI indicate an adequate degree of reproducibility for the production of fuel.

Table XVI. Precision of Pin-processing Data

Sample No.		Laboratory Data	Machine Mean Data (25 pins)	Machine Standard Deviation	Machine Variance
4	<u>Weight (g):</u>	68.7689	68.7645	0.010	0.0001
15		69.0009	68.9435	0.014	0.0002
4	<u>Length (in.):</u>	14.209	14.2088	0.007	0.00005
15		14.205	14.201	0.005	0.00002
4	<u>Volume (cc):</u>	3.8334	3.8348	0.01	0.00013
15		3.8554	3.8427	0.009	0.00008
4	<u>Density (g/cc):</u>	17.9394	18.0385	0.055	0.003
15		17.8972	18.0450	0.045	0.002

In addition to the calibration runs, a series of 23 pins were processed through the machine and then rechecked in the laboratory. As Table XVII shows, the data obtained by the machine deviate only slightly and randomly from the laboratory data.

Table XVII. Comparison of Fuel-Pin Data

Pin No.	Length (in.)			Weight (g)			Volume (cc)		
	Measured	Machine	Difference, mils	Measured	Machine	Difference	Measured	Machine	Difference
1	14.217	14.217	0	68.791	68.77	-0.021	3.812	3.807	-0.005
2	14.215	14.219	+4	68.521	68.51	-0.011	3.794	3.774	-0.020
3	14.212	14.221	+9	68.881	69.00	+0.12	3.814	3.811	-0.003
7	14.218	14.212	-6	68.63	68.62	-0.01	3.801	3.796	-0.005
9	14.218	14.219	+1	68.66	68.62	-0.01	3.801	3.813	+0.012
12	14.218	14.217	-1	68.48	68.48	0	3.792	3.783	-0.009
14	14.216	14.215	-1	68.40	68.49	+0.09	3.790	3.789	-0.001
17	14.211	14.215	+4	68.30	68.23	-0.07	3.783	3.783	0
19	14.219	14.219	0	68.43	68.44	+0.01	3.788	3.809	+0.021
20	14.213	14.214	+1	68.48	68.49	+0.01	3.794	3.789	-0.005
21	14.213	14.209	-4	68.74	68.73	-0.01	3.809	3.797	-0.012
22	14.217	14.214	-3	68.86	68.84	-0.02	3.813	3.792	-0.022
23	14.218	14.216	-2	68.66	68.63	-0.03	3.802	3.800	-0.002
24	14.217	14.218	+1	68.29	67.99	-0.30	3.786	3.774	-0.012
25	14.214	14.215	+1	68.48	68.49	+0.01	3.793	3.834	+0.041
26	14.219	14.219	0	68.69	68.69	0	3.804	3.782	-0.022
27	14.215	14.212	-3	68.26	68.23	-0.03	3.779	3.764	-0.015
28	14.221	14.220	-1	68.57	68.57	0	3.795	3.798	+0.003
30	14.212	14.209	-3	68.59	68.58	-0.01	3.801	3.789	-0.012
31	14.217	14.222	+5	68.76	68.74	-0.02	3.812	3.804	-0.008
33	14.213	14.216	+3	68.54	68.53	-0.01	3.798	3.788	-0.01
34	14.220	14.218	-2	68.81	68.89	+0.08	3.813	3.803	-0.01
35	14.219	14.217	-2	68.58	68.57	-0.01	3.801	3.817	+0.016

E. FARET

1. General

The cell and equipment arrangement for fuel handling, core instrumentation, core disassembly and disposition, and related requirements, such as remote control operations, viewing, and accessibility, have been reviewed in order to promote the definitive Title II design and to effect final definition for Construction Package I requirements.

Title II work with the Architect-Engineer has involved the following:

a. Based on the results of gas-changing operations at the EBR-II Fuel Cycle Facility and of previously conducted small-scale tests and calculations, it has been decided that the inert gas system used to change gas in the cell will use a displacement technique rather than the vacuum system outlined in Title I.

b. The reactor vessel design concept is being reviewed particularly with regard to methods of vessel support and evaluation of potential problems involving thermal stress.

c. Preliminary piping and instrumentation diagrams for the major FARET liquid metal systems and the heating and ventilation systems have been reviewed and comments have been transmitted to the Architect.

d. First drafts of the heat exchanger procurement package (Package III) have been reviewed.

e. Progress prints of major sodium-system piping, principally in the vault, have been reviewed.

2. Safety Analysis

The reactor kinetics have been successfully programmed by Bechtel for studies of system performance. Results have been obtained for the following four cases:

(a) and (b). Main coolant loop scram with reactor scram 0 and 0.3 sec later, and (c) and (d) reactor scram followed by main coolant system coastdown to 10 percent sodium and air flow starting 0 and 0.3 sec later.

Of interest in the FARET safety evaluation is the study of a nuclear incident which may be initiated by the partial loss of coolant or severe local perturbation of coolant flow into fuel subassemblies. If the loss of heat transfer occurs for a time interval sufficiently long for the fuel to start melting, then a positive reactivity insertion will result from the gravitational collapse of the molten core.

In order to estimate the effects from an incident of this type, a constant, low, inverse power period was initially introduced in the small core and zoned core loadings. Typical results from the meltdown code (briefly described in Progress Report for December 1963, ANL-6810, p. 18) are presented in Figures 12-16. For these cases negative feedbacks introduced by fuel expansion and Doppler effect are assumed not to occur.

Figure 12. Inverse Power Period, Power, and Energy Vs. Time (No Doppler) for FARET Small Oxide Core

Figure 13. Power Density, Energy Density, Inverse Power Period, and Net Reactivity Insertion Vs. Time for FARET Zoned System (7.4 in., 13.7 in., 15.8 in.)

Figure 14. Power Density, Energy Density, Inverse Power Period, and Net Reactivity Insertion Vs. Time for FARET Zoned Core (7.4 in., 13.7 in., 15.8 in.)

Figure 15. Power Density, Energy Density, Inverse Power Period Vs. Time for FARET Zoned System (7.4 in., 13.7 in., 15.8 in.)

Figure 16. Power Density, Energy Density, Inverse Power Period Vs. Time (Test Zone Only) for FARET Zoned System (7.4 in., 13.7 in., 15.8 in.)

In the case of the small core (see Figure 12) the time required for 70% and 100% of the core to melt was found to be 15.5 and 17 msec, respectively. The initial power period introduced into the meltdown code was about 3 sec. The inverse power period attained from the gravitational collapse of the core is about 2 to $3 \times 10^{-3}/\mu\text{sec}$ at an input energy density of $2 \times 10^4 \text{ J/cc}$ of fuel. It is estimated that at this energy density the core would start to disassemble.

As with the small core, a constant inverse power period of $\alpha = 9 \times 10^{-6}/\mu\text{sec}$ was initially introduced into the zoned system. The time required for 100% of the core to be melted is about 23 msec (see Figure 13); the melting temperature was taken as that of the uranium metal driver region. The energy density of $2 \times 10^4 \text{ J/cc}$ of fuel was reached in 29 msec, and the inverse power period at this time is approximately $1 \times 10^{-3}/\mu\text{sec}$.

The two cases for which the melting temperature was taken to be that of uranium oxide are presented in Figures 14 and 15. The initial power periods were 0.55 and 1.7 sec, respectively. In both cases the

gravitational collapse of the molten core resulted in an inverse power period of less than $2 \times 10^{-3}/\mu\text{sec}$ at the energy input of about $2 \times 10^4 \text{ J/cc}$ of fuel.

In Figure 16, the results are presented for an incident in which only the fuel in the test zone of the zoned system had collapsed. The time required for complete melting of the fuel was about 55 msec. The initial inverse period of $4 \times 10^{-6}/\mu\text{sec}$ was increased to about $1 \times 10^{-3}/\mu\text{sec}$ at an energy density of $2 \times 10^4 \text{ J/cc}$ of fuel.

These preliminary results indicate that the inverse power periods associated with loss of heat transfer and subsequent collapse of molten core are lower than had been anticipated. The lower inverse periods would result in lower energy yields during an excursion.

3. Component Development

a. Control Drive Mechanisms. The design of the reactor vessel cover penetration seal for the control rod drive appears to be a critical problem, for the choice of seal design dictates the arrangement of the drive components and the overall height of the control rod drive. To be acceptable the seal must fulfill the two main design parameters for the FARET reactor. (a) the seal must effectively maintain the integrity of the reactor vessel gas blanket, and (b) the seal must permit unrestricted movement of the drive shaft during scram. Two seal types have been chosen for test: a "sliding seal type" and a "bellows type" (see Figures 17 and 18).

The sliding seal design actually consists of two seals: an "O" ring static seal and a standard packing-type seal. The movement of the control rod shaft during normal operation is through the packing seal. The static seal is only broken during scram, thereby opening the gas blanket system for the reactor vessel for a very brief period (less than 0.5 sec). This is not considered objectionable. The "bellows type" seal utilizes a flexible metallic corrugated section between the control rod drive and the reactor vessel cover. Current design permits the bellows to undergo a 3-ft stroke during normal operation as well as for scram.

The seal-testing equipment, which will permit the evaluation of the seal designs at reactor vessel conditions (sodium at 1200°F) has been completed. Detailed design of the seal components is in progress.

The scram latch associated with the control rod drive is required to delatch in a flawless manner upon a scram signal and within acceptable release times. A simple lifting electromagnet is thought to be reliable for this purpose due to the lack of linkages for its operation. A magnet has been designed and is in the process of fabrication. This magnet

will be tested for its lifting capabilities and load-release time. Since there is a size limitation for the magnet, especially for the sliding seal-type drive, Armco iron capable of high flux densities was selected for the magnetic material.

Figure 17. Sliding Seal-type Drive

Figure 18. Bellows-type Drive

b. Reactor Head Assembly. A study has indicated the desirability that the head assembly be removed vertically through a ceiling access port. Such a scheme (see Figure 19) would permit access to control drive components for replacement or maintenance.

A four-point adjustable fixture with a load cell (dynamometer) and a positive connection to the reactor head and the crane located above the access plug are planned for raising the head assembly through the access port in the ceiling. Provisions are to be made for guiding the head of the reactor spool piece and for viewing facilities to observe the lifting and lowering of the cover.

Preliminary layouts show that the removal of other components through the access port may be difficult. A movable plug with inflatable seals located on the top of the ceiling, rather than having the plug under the ceiling, is being considered because of easier installation and alignment.

Figure 19. Proposal for Removal of Reactor Head Assembly

The desirability of an additional cell (room) located on the top of the reactor cell ceiling is being investigated, as well as a tank closure. The tank-type closure would be acceptable initially for tolerable low-level activity when full access to the maintenance cell may be permitted for manual maintenance. For more intense contamination, it is anticipated that considerable decontamination may be done within the tank to perform manual maintenance.

c. Wet Fuel Storage Cover. A scheme for using Tychoway* anti-friction way bearings to support the rotating storage cover, together with equally spaced roller bearings for resisting radial loading of the storage cover, is being studied. If successful, this would result in a substantial savings as compared with the cost of a large two-point contact ball bearing.

The application of a silicone-dacron inflatable-type seal for sealing of the argon gas blanket within the storage vessel from the reactor cell atmosphere is being investigated.

The required number and spacing of the fuel-storage tubes within the vessel is being determined. Following this the diameter of the fuel-storage vessel will be established.

4. CP-5 In-pile Experiment

The thermocouples for the heat flow transducer that failed to meet ANL specifications (see Progress Report for February 1964, ANL-6860, p. 38) have been replaced by the manufacturer; all except one of the 27 replaced thermocouples have been accepted.

An electric schematic and the wiring diagrams have been completed. The wiring of the auxiliary relay panel has started. Most of the electrical equipment has been received and only a few items are still to be delivered.

The constants for the valves in the control system have been calculated, and the valving arrangement for the two vacuum systems has been finalized. A special leak valve, consisting of a flattened copper coil wrapped on a shaft, has been automated and operated as a position-control system. (Flow through the tube is a function of shaft rotation.) After about an 8-hr operation, the coil developed a break. A new variable leak will be fabricated. After the new valve is available, the entire vacuum system will be simulated and the vacuum control system performance determined.

5. In-core Instrumentation

The FARET experimental program requires special instruments, particularly for the measurement of fuel-pin and coolant temperatures, and

*Scully Jones Manufacturing Company.

neutron and gamma fluxes. Electrical cable and connectors capable of withstanding high temperatures up to 650°C will be used. To develop and test such instruments and accessories, suitable facilities to enable testing at 650°C are required. (The fuel pin thermocouples will require test temperatures as high as 2760°C.) A suitable facility, approximately of 5-in. ID and 15 ft long, of which 10 ft is heated to produce an acceptable temperature profile, is planned.

The design of the facility is nearing completion, and fabrication is anticipated for next month.

6. Fuel Development

Two sets of uranium monocarbide pellets have been machined. Two additional holes, 0.18 cm in diameter, were placed on a 0.79-cm-diameter circle, in a straight line across the diameter of the pellet, to accommodate thermocouples for thermoconductivity measurements.

The two sets differed in pellet density. One set, having large-grain pellets, had an average density 74.5% of theoretical; the other, made with fine-grained material, had an average density 81.2% of theoretical. Each set was made up of 8 sections, of which 6 sections were approximately 1.96 cm in length and the other 2 sections were 0.94 cm long.

Chemical analyses of the uranium carbide obtained from United Nuclear, indicated that the -325 mesh* material contained 4.6 w/o carbon, whereas the -18 +80 mesh fraction contained 4.45 w/o carbon. This difference in carbon content may be the reason why the coarse fraction is difficult to crush.

In order to improve sintering, two techniques were tried. In the first, uranium carbide was heated in flowing argon to 1900°C; in the second the uranium carbide was mixed with carbon black to adjust its carbon content to 4.8-4.9% and then heated to 1900°C in flowing argon.

Pellets were fabricated from the uranium carbide heated to 1900°C by cold compaction at 25,000 psi and sintering in vacuum at 1850°C. These pellets were found to have an average geometric density of 85% of theoretical. Pellets fabricated from the untreated uranium carbide had an average geometric density of 75% of theoretical.

*United States Bureau of Standards mesh.

III. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. High-conversion Critical Experiment

The core in the High Conversion (Hi-C) Critical Experimental Facility has an atom ratio (H:U²³⁸) of 1.33, and a volume ratio of (H₂O/UO₂) of 0.44. It is fueled with 3 w/o enriched UO₂ as 0.94-cm-diameter pellets loaded into 1.06-cm-diameter stainless steel or 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 and has 4910 aluminum-clad fuel elements.

Activation traverses in the core with aluminum-clad fuel indicated a critical buckling of $(24.4 \pm 0.1) \times 10^{-4} \text{ cm}^{-2}$. Radial reflector savings of 10.7 ± 0.2 cm and axial reflector savings of 21.2 ± 0.2 cm were obtained by fitting Bessel and cosine functions to the radial and axial traverses, respectively, by means of computer computations.

Cadmium ratio (Cd R) measurements were made with gold, copper, dysprosium-aluminum, indium, and manganese foils, and 0.005-cm-thick aluminum or cadmium covers. The observed and interpolated values for the cadmium ratios are shown in Table XVIII.

Table XVIII. Cadmium Ratios

<u>Fuel</u>	<u>Thickness, cm</u>	<u>Observed Measurement</u>	<u>Interpolated Values</u>
Gold	0.005	-1.22 ± 0.01	1.22 ± 0.03
Copper	0.005	-1.80 ± 0.03	1.78 ± 0.01
3.7% Dy-96.3% Al	0.023	-9.18 ± 0.03	9.1 ± 0.1
Indium	0.003	-1.26 ± 0.01	1.25 ± 0.03

The interpolated cadmium ratio values were obtained from linear plots of Cd R versus the ratio of the number of hydrogen atoms per thermal barn of absorber present in the fuel elements for all the Hi-C cores. The linear relationship appears to fit the data closely over a wide range of core compositions. Further study is planned since there is hope that a systematic relationship may be shown which will simplify future work with UO₂-H₂O cores.

Careful measurements of the increase in U²³⁸ episcadmium activation at end gaps between Hi-C pellets have confirmed earlier measurements (see Progress Report for January 1964, ANL-6840, p. 52). A linear increase was obtained in the Np²³⁹ yield (determined by coincidence counting)

in depleted uranium metal foils irradiated in gaps of various widths. The increase was roughly 1.7% per 0.001 in. (67%/mm) with aluminum foil spacers (placed between depleted uranium metal foil and fuel) of the following thicknesses: 0, 1, 2, 4, and 8 x 0.001 in. The pellets were polished, aligned with foils, and rotated during irradiation on an 8-in.-diameter wheel with cadmium cover, in the ATSR graphite thermal column. Microscopic examination of the alignment indicated it was within 0.002-in. A least-squares fit of a straight line to the data gave a slope of 1.7 ± 0.1 per 0.001-in. gap thickness. Estimates based on rough calculations were twice as large as the measured effect.

Stainless steel-clad fuel will be installed in the central zone during the next month. Measurements of U^{238} to U^{235} fission ratio, δ^{28} , fission cadmium ratio in U^{235} , and thermal disadvantage factor will be made.

2. Subnanosecond RC Risetimes from Ionization Counters

The measurement of fast pulses by means of gas-ionization counters and proportional and semiconductor counters in a current mode has recently become feasible with the advent of transistorized current amplifiers having high-gain bandwidth, sufficiently low noise, and adequate stability.⁴ Current, in contrast with voltage, provides exceptionally fast low-jitter pulses quite suitable for coincidence counting in the nanosecond and picosecond regimes. With current amplification, one obtains large signals from small resistive inputs, a pulse height which is not dependent on track origin, and a risetime which, aside from being restricted by the amplifier bandwidth, is only limited by the small RC input or by the charge induction time (including nuclear track formation time). The discharge time of the capacity representing the chamber and the amplifier input determines the speed of rise of the induced current pulse. If a uniform field and constant drift velocity may be assumed, the current pulse promptly reaches its maximum, remaining constant as long as the electron sheath is clear of the collection electrode. For an ionization track parallel to the chamber plates, the electron swarm will reach the collecting electrode simultaneously, resulting in a pulse width essentially equal to the transit time of the negative charge carriers across the chamber.

In a typical pulse profile (see Fig. 20) observe that the pulse width is a little over 20 nsec and the risetime around 3 nsec. The width⁵ is consistent with a drift velocity for electrons approaching 10^7 cm/sec.

⁴C. Rush, New Techniques for the Designing of Fast Rise Transistor Pulse Amplifiers, Rev. Sci. Instr. 35, 149-156 (1964).

⁵Arthur H. Snell (Ed.), Nuclear Instruments and Their Uses, John Wiley & Sons, New York (1962).

Figure 20

A Typical Oscilloscope Pulse Profile from a Cf^{252} Fission Fragment Shown on the Oscilloscope in Conjunction with a Methane Flow Ionization Counter and a Transistorized Amplifier Using the Current Mode of Operation.

Figure 21 provides a schematic of the test arrangement. The ionization counter output was obtained under the following conditions:

- (1) A thin deposit of spontaneously fissioning Cf^{252} is placed on one electrode of a parallel plate counter.
- (2) Plate spacing is 1-2 mm; bias voltage, +300 V.
- (3) Filler gas is 99% methane at about 1 atm in a flow-type arrangement.
- (4) The collector electrode is connected to the emitter of the input stage of the current amplifier.
- (5) Amplifier gain is 1600, and risetime around 3 nsec; note that this is about the risetime of the pulse trace shown in Figure 20.
- (6) A Tektronik 519 oscilloscope with risetime of 0.3 nsec is used; vertical sensitivity, 10 V/cm; and input impedance of 125 Ω .

Figure 21. Schematic Arrangement of Test Equipment

By driving directly into the low impedance of the amplifier, the effect of stray capacitances is minimized to the point at which counter risetime is normally negligible with respect to the amplifier. With a chamber

capacity of 5 pF, the addition of 5pF for stray capacity, and an apparent input impedance of $10\ \Omega$ that may be presented by this particular amplifier, an RC time of 100 psec is computed. Since, at present, the amplifier limits upper frequency response, much larger counter capacities may be tolerated while still maintaining fast risetimes.

Figure 22 demonstrates the effect of inserting a $40\text{-}\Omega$ resistor to obtain an input impedance of $50\ \Omega$ and then coupling the counter to the amplifier with almost 40 ft of RG/58 cable. Since this coaxial cable does not have an especially good transmission bandwidth, some degradation of risetime is noticeable.

These remarks, with suitable qualifications, also apply to semiconductor detectors. Figure 23 indicates the pulse obtained from a 300-ohm-cm junction counter based on phosphorus diffusion into p-type silicon. The pulse width is about 7 nsec, and again the risetime is a function of the amplifier performance. As long as operation may be restricted to the depletion depth, the current output from solid-state counters is larger than that available from gas-ionization counters.

Figure 22. A Typical Oscilloscope Pulse Profile from a Cf^{252} Fission Fragment Showing Degradation of Risetime Caused by Interposing 40 ft of RG/58 Cable between the Methane Flow Ionization Counter and the Transistorized Amplifier Using Current Mode of Operation.

Figure 23. A Typical Oscilloscope Pulse Profile from Am^{241} Decay Fragment Using a Semiconductor Detector (+100 V Bias) and a Transistorized Amplifier Using the Current Mode of Operation.

B. Theoretical Reactor Physics

1. Fast Reactor Studies

The ELMOE code⁶ is often employed in correlation studies of calculated and experimental masses of ZPR-III fast critical assemblies to obtain the effective group cross sections for transport and for elastic transfer for the cores. The critical masses are then usually calculated

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

by use of the DSN neutron transport program⁷ in the S_4 approximation. For these studies the ELMOE averaging has generally been in the P-1 approximation. As the ELMOE program also has consistent P-1 and consistent B-1 options, it was of interest to calculate critical volumes of a few ZPR-III assemblies with the use of effective cross sections obtained by the consistent B-1 option. Four assemblies: No. 14, with graphite diluent; No. 22, with U^{238} diluent; No. 23, with aluminum diluent; and No. 32, with steel diluent, were studied.

The calculated ratios of critical volumes by means of the consistent B-1 averaged cross sections relative to the critical volumes with the simple (nonconsistent) P-1 averaged cross sections were 0.991, 0.993, 0.984, and 0.951, respectively. The ratios correspond to increased calculated reactivities of +0.2% k, +0.1% k, +0.4% k, and +1.2% k, respectively. It may be noted that the ZPR-III calculations already tend to overestimate the reactivities and the use of consistent B-1 averagings rather than simple P-1 averagings tend to increase further the overprediction of reactivity.

2. Studies of the Sodium Void Effect

Work reported at the Breeding and Safety Conference⁸ at Argonne has been extended, with results as given in Table XIX. It is now definite that the spectral and capture components of the sodium reactivity effect, for removal from a uniform core, are only weakly dependent on core geometry but depend mainly on core composition. The leakage component is, of course, strongly dependent on geometry and is not very sensitive to changes in the isotopic content of the fuel, in this case to the replacement of U^{238} with Pu^{240} .

Table XIX. Components for Void Effect in % δk with Loss of One-half of Sodium Volume; PuO_2 - UO_2 Fuel, Stainless Steel Structure

Sphere Volume, liters	Sodium, v/o	Approx. $U^{238} + Pu^{240}$ Pu^{239}	Leakage		Inelastic		Elastic		Capture ^(b)		Spectral + Capture	
			Sphere	Infinite Slab ^(c)	No Pu^{240}	With Pu^{240} ^(a) + FP	No Pu^{240}	With Pu^{240} ^(a) + FP	No Pu^{240}	With Pu^{240} ^(a) + FP	No Pu^{240}	With Pu^{240} ^(a) + FP
1000	52	5.8	-1.68	-0.69	0.33	0.50	0.44	0.89	0.07	0.04	0.84	1.43
2000	52	7.0	-1.39	-0.69	0.43	0.60	0.77	1.20	0.08	0.04	1.28	1.84
3000	52	7.6	-1.12	-	0.48	0.64	0.92	1.31	0.08	0.04	1.40	1.99
2000	59	5.4	-2.21	-	0.42	0.63	0.52	1.22	0.12	0.07	0.94	1.92
3000	59	5.9	-1.89	-	0.50	0.70	0.80	1.39	0.13	0.07	1.37	2.16
5000	59	6.6	-1.58	-	0.56	0.80	1.06	1.68	0.13	0.07	1.75	2.55
2500	70	4.7	-2.85	-1.16	0.45	0.78	0.40	1.19	0.15	0.08	0.95	2.05
5000	70	5.7	-2.47	-1.23	0.68	0.97	0.92	1.75	0.18	0.10	1.60	2.82

^(a) Pu^{240} replaces U^{238} ; $Pu^{240}/Pu^{239} = 0.4$. Fission products (FP), 5 a/o (2.5% burnup).

^(b) Based on g Γ of 2.85-keV sodium resonance = 0.27 eV.

^(c) Core composition same as for sphere.

⁷B. Carlson et al., The DSN and TDC Neutron Transport Codes, LAMS-2346 (1960).

⁸H. H. Hummel, K. E. Phillips, and A. L. Rago, Calculations of Sodium Void Reactivity Effect for Large Fast Oxide Reactors in Spherical and Slab Geometry, Proc. Conf. on Breeding, Economics, and Safety in Large Fast Power Reactors, Argonne National Laboratory, October 7-10, 1963, ANL-6792 (Dec 1963), pp. 65-74.

Although the leakage component for a sphere varies significantly with size, that of the slab is nearly independent of thickness over a considerable range. This is due to the increasing effectiveness of the reflector as slab thickness decreases,⁹ and limits the usefulness of a small L/D ratio in combating the sodium coefficient problem. It is evident from the data in Table XIX that a much higher enrichment is required for an infinite slab than for a sphere to achieve a zero effect for removal of sodium from the core only.

C. High-temperature Materials Development

1. Zirconium Alloys

a. Corrosion in Superheated Steam

(i) Alloy Modification. Zirconium alloys that are corrosion resistant in superheated steam are being modified to reduce surface metal embrittlement associated with oxygen absorption resulting from exposure to steam at 650°C. This is being sought by controlled immobilization of the metal structure through alloying with Be, Ge, Si, Al, Mg, or Nb, which were chosen for their low neutron-absorption cross sections and for their known or suspected ability to harden or strengthen zirconium. To date, twenty alloys containing silicon or beryllium have been prepared in the 50 to 75 a/o range, half of which were based on crystal-bar zirconium and half on a master alloy of Zr- 1 w/o Cu-1 w/o Fe. It is expected that usable alloys will contain lower amounts of the stabilizing addition, consistent with ductility requirements. However the corrosion, hardness, and behavior with respect to oxygen absorption of the more concentrated alloys will be of immediate interest. Simple mechanical tests have verified the anticipated brittleness of the silicon alloys and have revealed extreme toughness in the beryllium alloys before ultimate brittle fracture.

(ii) Low-pressure Corrosion Tests. Zirconium alloys containing copper or nickel and iron, which are resistant to corrosion in steam at 650°C and 42.2 kg/cm², have been tested at the same temperature and 1 torr water-vapor pressure for a total of 12.8 days after initial exposure to steam at the higher pressure for 0.9 day. Although the sample weight gains observed were somewhat lower than those in tests at higher pressure, the rates of gain after 5 days were essentially the same as at 42.2 kg/cm². Lower total gains may have been due to a test temperature significantly lower than 650°C, which is now known to have prevailed during the first low-pressure exposure period (2.7 days), or possibly to a real effect of low pressure on the initial corrosion behavior. The rate results reveal the lack of pressure dependence of the corrosion reaction (at least after the initial exposures) and give additional interest to the results of electrical measurements made in the low-pressure environment.

⁹W. G. Davey and J. M. Chaumont (unpublished).

(iii) Electrical Measurements. Additional electrical measurements are being obtained with a flat platinum probe spaced $50\ \mu$ from a corroding zirconium alloy sample in 0.1 to 10 torr water-vapor pressure environment at 540 to 750°C. The gap resistance to passage of 100-cycle AC current has been measured as a function of pressure at various temperatures. Considerable carrier emission occurred above about 650°C, and a pressure increase to the 10-torr range decreased the mean free path of the carriers, interfering markedly with the conductance of the gap. The carrier species have not been identified, but the appearance of the wave shape as pressure is varied at the higher temperatures (e.g., 700°C) suggests the presence of both electrons and gas ions. These effects are of interest in relation to a hypothetical hydriding mechanism, which explains the previously observed behavior with respect to hydrogen absorption of these materials during corrosion in steam, i.e., the metal absorbs a lower percentage of corrosion product hydrogen at 650°C than at 540°C (the steam pressure using 42.2 kg/cm² for both temperatures) because of increased thermionic emission activity at the higher temperature, which promotes hydrogen ion discharge, perhaps just outside of the corrosion film surface. The relevance of the results to the oxidation mechanism is being considered and the observations are continuing.

2. Uranium Carbide and Uranium-Plutonium Carbide

a. Uranium Carbide. A sintering evaluation was made of samples of uranium monocarbide, designated as Run UC-2, prepared by precipitation from a zinc-magnesium melt. The uranium carbide contained 5.4 w/o carbon, 0.39 w/o oxygen, and 380 ppm nitrogen. According to X-ray analysis, the only crystalline phase present was uranium monocarbide.

The material as received was pressed with 1% binder into 0.952-cm-diameter pellets and vacuum sintered at 1600°C for 3 hr. Additional material was ball milled for 2 hr and processed as above. The glovebox atmosphere in which the milling and pressing were done contained from 200 to 300 ppm water.

Geometric densities were determined for the sintered specimens, and the unit cell sizes were obtained by Nelson-Riley extrapolations (see Table XX).

b. Irradiation of Fast Reactor Fuels. One phase of the overall irradiation program involves transient irradiation tests in TREAT of mixed UC-20 w/o PuC and PuC powders, vibratorily compacted in either Type 304 stainless steel or Nb-1 w/o Zr tubing. The initial two experiments, which provided data on the energy release in PuC as a function of integrated TREAT reactor power, have been analyzed. The energy release in a specimen with a 0.13-mm (0.005-in.) cadmium cover was 1.10 cal/g plutonium per MW sec of total reactor power. The energy release from

a specimen without the cadmium cover was $3.42 \text{ (cal)/(g-Pu-MW sec)}$. The plutonium in both cases contained 95% Pu^{239} . Normalization of the data to the infinitely dilute case (in which there is no self-shielding) without the cadmium cover yielded an energy release of $40.1 \text{ (cal)/(g Pu-MW sec)}$.

Table XX. Data for Sintered Specimens

	Average Geometric Density (% T.D.)*	Unit Cell Size ($\text{\AA}$)
As Received	61.2	4.9598
Ball Milled**	85.0	4.9608

*T.D. = Theoretical Density.

**A polished and etched specimen of the ball-milled material showed a central core region containing three phases: UC, UC_2 , and UO_2 , and the periphery composed of UC with traces of UO_2 . No UO_2 was present in this outer portion.

3. Thermal Properties of Uranium Monosulfide

A heat-pulse apparatus for measuring specific heats and thermal diffusivities was calibrated at room temperature for some common metals. The thermal conductivities calculated from these properties were within 5% of the best literature values.

The thermal diffusivity (α) and the heat capacity (C_p) of 86% dense uranium monosulfide was determined as follows:

<u>Specimen Thickness (cm)</u>	<u>$\alpha \text{ (cm}^2 \text{ sec}^{-1}\text{)}$</u>
0.101	0.038
0.1525	0.036
0.196	0.040

The specific heat (C_p) of the 0.101-cm-thick specimen was determined by comparing its temperature increase upon flashing with that of copper and Armco iron:

<u>Metal Standard</u>	<u>Uranium Monosulfide $C_p \text{ (cal gm}^{-1} \text{ }^\circ\text{C}^{-1}\text{)}$</u>
Copper	0.048
Armco Iron	0.046

The average value of $0.047 \text{ cal gm}^{-1} \text{ }^{\circ}\text{C}^{-1}$ is in good agreement with that reported by Westrum et al.,¹⁰ of $0.045 \text{ cal gm}^{-1} \text{ }^{\circ}\text{C}^{-1}$ at 25°C .

D. Other Reactor Fuels and Materials Development

1. Urania Burnable Poisons

In nuclear reactors, "poisons" are materials with such high absorption cross sections for neutrons that they limit the number of neutrons available for fission. Poisons are therefore often used to control nuclear reactors. As the nuclear fuels are burned up, the reactivity may decrease excessively unless the poisons also burn up at a compensating rate. Such poisons, called "burnable poisons," and are sometimes used with fuel materials, such as urania, to obtain a more nearly uniform rate of power output. Since gadolinium is being considered for use as a burnable poison, the effects of gadolinia additions in urania are being studied.

A face-centered cubic solid solution is formed by the addition of small amounts of gadolinia to uranium dioxide when sintered in hydrogen or in argon. Considerably more gadolinia is needed to form a solid solution in air-sintered compacts of U_3O_8 and gadolinia.

X-ray analysis of air-sintered compacts of U_3O_8 and $\text{GdO}_{1.5}$ indicated that with 5 m/o $\text{GdO}_{1.5}$, the formation of the fluorite-type structure had begun. The predominant crystallographic pattern was that of U_3O_8 , however. With the additions of 10 and 20 m/o $\text{GdO}_{1.5}$, the amount of solid solution increased while the U_3O_8 content decreased. With the addition of 30 m/o $\text{GdO}_{1.5}$, no free U_3O_8 remained. Diffraction patterns indicated only the existence of the solid solution until 70 m/o $\text{GdO}_{1.5}$ was present. On the diffraction pattern for 30 m/o U_3O_8 -70 m/o $\text{GdO}_{1.5}$, lines corresponding to high-temperature-sintered gadolinia were evident. Subsequent additions of gadolinia further decreased the intensity and clarity of the diffraction lines for the solid solution while increasing the intensity of the lines for gadolinia.

Lattice parameters of the solid solution were relatively uniform at $5.405 \text{ \AA}$ until the addition of 30 m/o $\text{GdO}_{1.5}$. At higher gadolinia contents, the parameters decreased linearly through the 60 m/o $\text{GdO}_{1.5}$ addition (parameter of $5.382 \text{ \AA}$).

2. Nondestructive Testing

a. Neutron Radiography. High-resolution neutron radiographs of a number of alloys containing materials having high neutron attenuation

¹⁰Westrum, E. F., Jr., and Gronvold, F., "Chemical Thermodynamics of the Actinide Element Chalcogenides," Thermodynamics of Nuclear Materials, IAEA, pp. 3-37 (1962).

are being taken as part of a study initiated by A. A. Fahmy (ISE). Neutron radiographs of B-Al, Cd-Pb, and Cd-Bi alloys, yielding resolution sufficient for 100-times enlargement, are now under study.

A neutron radiographic method for comparing the neutron attenuation of experimental reactor control materials with that of cadmium has undergone preliminary tests. The predicted accuracy appears to be better than 10 percent. Comparison data for a number of samples in both the thermal and resonance neutron-energy regions are now being obtained.

The possibility of employing image intensifier methods for neutron image detection has also been under investigation. Preliminary tests with a standard vacuum tube, photoemissive type of X-ray intensifier* indicated no appreciable response to neutrons either directly or with a converter plate such as cadmium or gadolinium on the face plate of tube. This result was not entirely unexpected since the radiation emitted from the converter material would have to penetrate the glass face plate before reaching the input phosphor layer of the intensifier tube.

Further studies involving the relative light output of one of these input phosphor screens did indicate that increased light was obtained in a given neutron intensity when the screen was backed directly with either cadmium or gadolinium as compared to the light from the phosphor alone. Other phosphors and phosphor-converter material combinations are now under study in an attempt to find a more promising cathode material for a possible neutron image intensifier.

b. Correlation of Heat Transfer and Bond Quality. The correlation between ultrasonic transmission capabilities of various types of mechanical bonds and their heat transfer capabilities is being studied. To determine the thermal properties of the bonds the thermal pulse method for measuring diffusivity and conductivity developed by Parker et al.,¹¹ is being utilized. The front surface of the specimens are uniformly irradiated by a very short (approximately 600 μ sec) pulse of radiant energy supplied by a xenon flash tube. The temperature history of the back surface was continuously monitored by feeding the output of a thermocouple in contact with the back surface into a differential amplifying system and then into an oscilloscope.

Diffusivity and conductivity measurements are continuing for the copper braze-bonded specimens, and it is expected that the thermal measurements on this set of specimens will be completed in a month.

*An X-ray image intensifier, Rauland Type R-6175-RP, kindly loaned to us by the Rauland Corporation, was used for these tests.

¹¹Parker, W. J., Jenkins, R. J., Butler, C. P., and Abbott, G. L., Flash Method of Determining Thermal Diffusivity, Heat Capacity and Thermal Conductivity, J. Appl. Phys 32(9), 1679 (1961).

The system is being recalibrated, for the aging of the flash tube has made a small reduction in its efficiency. This results in a reduction in Q , the amount of heat absorbed at the front surface of the specimen.

c. Application of Infrared Radiation to Nondestructive Testing.

Investigation of the possible use of infrared in nondestructive testing has continued. The gray-scale meter for the infrared imaging system had too low an input impedance for the oscilloscope CRT. A high-input impedance transistorized circuit for the meter was built but was not satisfactory because of thermal drift. A stable vacuum tube circuit is now being constructed which should solve this problem.

Investigation of the use of a thermographic phosphor screen as an infrared image converter has been resumed.

d. Ultrasonic Propagation in Liquid Metals. An improved transducer assembly for ultrasonic studies in liquid sodium has produced good ultrasonic echoes from the bottom of a sodium tank through a sodium depth of greater than 40 cm. The reflected pulse still has appreciable width (greater than 50 μ sec) and further attempts to reduce this pulse width are now under way.

E. Remote Control Engineering Development

1. Viewing Systems

Glasses containing small amounts of cerium or other additives are expected to provide improved radiation resistance to induced coloration and other effects.

The base glass used in the current studies is a potassium aluminoborate glass, of $\text{Al}_2\text{O}_3 \cdot 4.5 \text{ B}_2\text{O}_3 \cdot \text{K}_2\text{O}$, which has a good optical transmission in the ultraviolet and is easy to melt. When cerium is added and the glass is melted under oxidizing conditions both Ce^{3+} and Ce^{4+} ions are present; when melted under reducing conditions, most of the cerium is in the Ce^{3+} state. Comparison of the absorption spectra of Ce^{3+} in glass with those in solutions and crystals leads to a conclusion that Ce^{3+} probably exists in two different structural configurations in which the coordination number, i.e., the number of oxygen atoms immediately surrounding the cerium, is different. The Ce^{4+} ions may also exist in two coordinations.

When the glass is irradiated, Ce^{3+} acts as a hole trap and Ce^{4+} as an electron trap. In this glass the Ce^{3+} ions prevent the formation of centers with absorption bands in the visible region, but they do not prevent the growth of some centers with absorption bands in the ultraviolet. Whether the cerium ions in different coordinations have different capture cross

sections is not known at present. The difficulty in analyzing the results lies in the fact that the absorption spectra of cerium in its different valence states and coordinations overlap one another.

Other additives to the potassium aluminoborate glass which have been studied include thallium, copper, lead, and silver in molar concentrations ranging from 0.0005 to 0.05. It was found that the capture cross sections of these elements for electrons and holes are generally less than those of cerium. In addition, unlike cerium, some of the induced valence states absorb in the visible region. In the case of silver-doped glass, the silver enters the glass as an Ag^+ ion with an absorption band in the ultraviolet. The Ag^+ ion acts as both a hole trap and electron trap. On irradiation one gets Ag^0 atoms and Ag^{2+} ions, but unfortunately, the absorption bands associated with Ag^0 atoms extend into the visible part of the spectra. Further studies of this type are in progress.

F. Heat Engineering

1. Boiling Sodium Heat Transfer Facility

a. Niobium-zirconium Loop Construction. Brazing of the cooling lines to the vacuum chamber continues at the vendor's plant. Some loss in time occurred when an improperly annealed dished head cracked during the brazing and had to be replaced. The diffusion pump has been obtained, and the oven is essentially complete.

The circuit diagram for the vacuum-chamber controls has been approved. A universal attachment for use in lifting the various portions of the chamber and its oven has been designed, and some spare parts have been ordered. A beginning has been made on the planning of utility services for the vacuum chamber and related equipment. It is hoped that vacuum chamber power can be obtained from a circuit which is independent of other experiments in the building.

Circuitry for the 150-kW power-supply controller and power-measuring instruments has been studied, and some miscellaneous small parts associated with this supply have been ordered.

b. Heater Experiments. The loop enclosure containing the radiantly heated forced-convection stainless steel loop has been modified so that it may be used as a vacuum chamber. Although evacuation to 0.20 in. Hg absolute has been already obtained, refinements are expected to provide a vacuum of approximately 10^{-3} torr.

The radiantly heated section is complete and will be welded into the system when refinements on the vacuum enclosure are completed. A full-sized model of the tantalum radiant heating element has performed satisfactorily at temperatures around 2000°F.

Drawings for the small-scale electron bombardment heating experiment have been released for fabrication.

2. Heat Transfer in Double-pipe Heat Exchangers

The two region Sturm-Liouville problem (see Progress Report for January 1964, ANL-6840, p. 68) has been applied to the case of cocurrent laminar flow in concentric-tube heat exchangers. Values of fully developed Nusselt numbers and of a new quantity, called "the effectiveness coefficient," were obtained as functions of the heat exchanger operating conditions. Use of these makes possible a computation of the heat exchanger effectiveness when the customary heat exchanger design relations are not truly applicable. The results of this study have been described in detail in a paper, entitled The Graetz Problem in Cocurrent Flow Double-pipe Heat Exchangers, submitted for presentation at the Seventh National Heat Transfer Conference.

Computations for laminar flow based on a perturbation type of analysis with plug flow representing the "unperturbed" case have been made with an IBM 704, but were not considered to be completely satisfactory, mainly because of the relatively large amount of machine time required. As a result, and because various extensions are desirable, the problem has been formulated for solution by analogue computer.

Work has begun on the design of an experiment to verify the results predicted previously for the turbulent flow of liquid metals in cocurrent-flow double-pipe heat exchangers (see Monthly Report for October 1963, ANL-6801, p. 53). Temperature measurements will be made in mercury-to-mercury cocurrent-flow double-pipe heat exchangers.

3. 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. Transient Behavior Heat Transfer Loop. The unstable behavior of a heat transfer loop which occurs when the single-phase coolant approaches its thermodynamic critical point is being investigated. An experimental loop, designed to operate by either natural or forced convection, is being utilized.

A turbine-type flowmeter (Pottermeter) and insulated wall thermocouples have been successfully incorporated in the system. The Pottermeter was installed to check whether large differential pressure oscillations across a Venturi were a result of pressure waves crossing the Venturi pressure taps or an indication of flow fluctuations. Welding the thermocouples to the electrically heated test section had resulted in excessive "noise." A sheet of mica, 0.0007 in. thick, was used to insulate the thermocouples from the test section. This reduced the electrical noise to such an extent that the wall temperature can now be recorded on the Visicorder simultaneously with the other variables. Significant experimental results observed to date are:

1. During the relatively fast (about 15 cps) pressure oscillations encountered when the test-section bulk temperature is 10-100°F below the critical temperature, the Pottermeter does not indicate appreciable flow fluctuations. This contrasts with the rather large differential pressure oscillations across a Venturi which have previously been observed. (It is now believed that these are caused by acoustic-type pressure waves traveling past the Venturi pressure taps.)

2. When the oscillations are encountered (or when they are damping out), a large change in wall temperature (up to 50°F) is observed at some point in the heated test section.

3. When the test-section outlet bulk temperature approaches or exceeds the critical temperature, a different type of oscillation may occur. Both the pressure and velocity now vary regularly, although more slowly (0.05 cps). In this case the fluid bulk and the wall temperature also tend to vary in a periodic manner.

Future plans include the study of the acoustic-type pressure disturbances, whose magnitude under the proper condition may be several hundred ps_i. Similar pressure oscillations have been noted in the literature, both for two-phase and supercritical systems. In most cases they have been of higher frequency than for the present case, resulting in loud shrieks and "boiling songs." The frequency of the vibrations observed in this investigation have been below the audible range.

b. Two-phase Flow Characteristics during the Complete Vaporization of a Fluid. The required equipment is being constructed. Assembly of the test section will be undertaken shortly in order to perform pressure tests on the unit.

Power supplies for the lighting units are to be shipped on March 18. Thus, a test of the two-color light system on an air-water loop can be made in the near future.

It was previously reported (see Progress Report for February 1964, ANL-6860, p. 72) that an attempt was being made to correlate the viscosities of various Freons. At present, the saturated liquid viscosities of Freons and water have been obtained as a single expression in terms of the reduced pressure ratio for a pressure ratio range from 0.001 to 0.20. The simple relationship for the liquid viscosity is

$$\mu_f = \mu_{\text{ref}} C P_r^n,$$

where μ_f is the saturated liquid viscosity, μ_{ref} is the saturated liquid viscosity of the particular substance at a reduced pressure of 0.03, C is a constant ($\cong 0.4$), and the exponent n is approximately -0.262. The range of deviation of this expression from the available data is approximately 8 percent. The values listed above are tentative. A single expression for the pressure ratio range from 0.20 to 1.0 is presently being formulated. A similar treatment for the saturated vapor viscosity will be undertaken.

The surface tension of the Freons can be correlated over the reduced pressure range from 0.001 to 0.10 by a single expression:

$$\sigma = 7.9 P_r^m \text{ (dynes/cm),}$$

where m is approximately -0.19. Deviations increase at high pressure ratios. A better correlation of surface tension for higher pressure ratios for a range from 0.01 to 1.0 is

$$\sigma = \sigma_{\text{ref}} A \ln P_r,$$

where σ is the surface tension of the saturated liquid, σ_{ref} is the saturated liquid surface tension evaluated at a reduced pressure of 0.03, A is a constant (approximately -0.286), and P_r is the ratio of the saturation pressure to the critical pressure.

c. Propagation of Void Waves in an Air-Water System. Void measurements by three different methods were made during a series of runs on the natural-circulation air-water loop. The void fractions obtained by correcting the pressure drop readings for frictional losses agreed, within 5%, with those obtained by the gamma-ray attenuation method. Measurements with the electrical resistivity probe were, however, about 25% lower.

The average void fraction by the probe method was obtained by integrating the void profile, since the probe only measures local voids. Calibration of the probe method is not, at the present time, possible. The reason is that it is not known if the error is a function of pipe radius. In order to determine this, it is necessary to measure local voids by another method of which none are known. More experiments are being conducted in order to determine precisely the cause of the error.

All six channels of the Void Probe Trigger have been completely wired and checked. The Air Flow Regulator, the electronic unit necessary for producing a square-wave air input, has also been completed. The probes needed for the investigation of propagating void waves have been constructed.

G. Chemical Separations

1. Chemistry of Liquid Metals

Six intermetallic compounds are known to exist in the system cerium-cadmium: CeCd_{11} , CeCd_6 , $\text{Ce}_3\text{Cd}_{13}$, CeCd_3 , CeCd_2 , and CeCd . The equilibrium pressures of cadmium over the following two-phase regions are being measured: CeCd_{11} - CeCd_6 , CeCd_6 - $\text{Ce}_3\text{Cd}_{13}$, $\text{Ce}_3\text{Cd}_{13}$ - CeCd_3 , CeCd_3 - CeCd_2 , and CeCd_2 - CeCd . The temperature range studied thus far is 361-520°C. These data will be used in calculating the thermodynamic properties of these phases.

2. Fluidization and Volatility Separation Processes

a. Recovery of Uranium from Low-enrichment Ceramic Fuels

(i) Laboratory-scale Fluid-bed Fluorinations. Laboratory work in support of the pilot-plant operation is directed primarily towards the determination of the optimum conditions for the fluorination of U_3O_8 - PuO_2 mixtures in a fluidized bed of alumina granules and for the efficient removal of plutonium from the fluid bed. The magnitude of the plutonium loss to be expected upon discarding of the alumina to waste cannot yet be stated, since this will depend upon a number of factors not yet determined, such as the number of times that the alumina bed can be reused.

Recent fluid-bed experiments have been made to determine the effect of added fission product elements and uranium on plutonium retention on the alumina bed material. In these experiments, PuO_2 alone or in a powdered mixture with U_3O_8 or selected fission product element was injected into the fluidized alumina bed, where the material was contacted with fluorine. During the injection period, most of the uranium and about 50 w/o of the plutonium were converted to hexafluorides. Fluorine gas was then recycled through the alumina bed to remove the remaining plutonium. The temperature during the feeding-fluorination period was 450°C in tests involving U_3O_8 fluorination and 500°C in other tests. During the 10-hr recycle-fluorination period the temperature was 550°C.

The experiments showed that the quantity of plutonium retained on alumina appears to be determined by the quantity of plutonium fed to the reactor; a 7.5-fold increase in the quantity of plutonium fed to the reactor resulted in nearly a 20-fold increase in the residual plutonium concentration on the alumina bed. The plutonium concentration on the

alumina bed after fluorination is not affected by the presence of uranium or fission products in the feed mixture provided that U_3O_8 fluorination is conducted at 450°C .

(ii) Plutonium Pilot Plant Facility. Since the self-decomposition of PuF_6 to PuF_4 from alpha-radiation is known to occur (see Progress Report for January 1964, ANL-6840, p. 75), only short-term storage of the mixed UF_6 - PuF_6 fluorinator product is desirable. In the pilot plant facility, it is proposed that the mixed hexafluorides be immediately converted to stable oxides by reaction with hydrogen and steam. This conversion will be carried out in a 2-in.-diameter fluid-bed reactor to be installed in the pilot plant. In preparation for tests involving plutonium hexafluoride, engineering studies are being made on the conversion of UF_6 to dense UO_2 particles in a 3-in.-diameter fluid-bed reactor. Current work involves an investigation of an oxidation-reduction procedure for the fluid-bed preparation of seed particles as an alternative to the present method of mechanical grinding (see ANL-6840, p. 75). A run was successfully carried out with a fluid bed composed of a narrow size range of UO_2 particles (-20 +40 mesh; tapped bulk density, 5.6 g/cc).

b. Supporting Study

(i) Corrosion Tests of Nickel. Corrosion studies are being carried out in support of the fluidization fluoride volatility program, primarily to establish the suitability of nickel as a material of construction for both the high-enriched alloy fuel process and low-enriched ceramic fuel process (see Progress Report for December 1963, ANL-6810, p. 39). Weld specimens consisting of $\frac{3}{16}$ -in-thick nickel-200 plate welded with either nickel-200 wire as filler metal or nickel-61 weld filler metal were exposed for 480 hr to the following environments which simulated process gases:

- (1) 50 v/o F_2 in N_2 at 500°C ;
- (2) 50 v/o HCl in N_2 alternating with 50 v/o F_2 in N_2 every 24 hr at 450°C ;
- (3) 50 v/o F_2 in N_2 alternating with 50 v/o O_2 in N_2 every 24 hr at 500°C .

The rates of corrosion of the welded specimens made with nickel-200 filler in all environments were found to be significantly lower (2 to 5 mils/yr) than those found for welds made with nickel-61 filler metal (70 to 800 mils/yr). All welds were examined metallographically for evidence of intergranular penetration; none was found. The corrosion of nickel metal specimens exposed to the alternating gas mixtures was less than the sum of the corrosion expected by exposure to the separate gases. Although

these data indicate that nickel equipment welded with nickel-200 filler should be suitable for use under the proposed process conditions, a number of factors, such as the effect of thermal cycling, remain to be evaluated.

3. General Chemistry and Chemical Engineering

a. Preparation of Uranium Monocarbide. Work has begun on a method to produce uranium monocarbide by hydriding and dehydriding uranium metal to form powder and reacting the powdered metal with a methane-hydrogen mixture at 600-800°C in a fluidized bed. In preliminary work, one batch of uranium hydride (UH_3) powder was prepared and was fluidized in a transparent 1-in.-diameter column. The quality of fluidization was acceptable. A 1-in.-diameter stainless steel fluidizing column is being fabricated for use in further investigations.

b. Preparation of Plutonium Carbide-Uranium Carbide Solid Solution. Techniques and equipment that will be used in the preparation of plutonium carbide-uranium carbide solid solution have been tested in a preliminary run in which uranium monocarbide was prepared. Uranium metal was dissolved in zinc at approximately 630°C and was simultaneously oxidized to uranium chloride by zinc chloride and transferred to a salt phase (MgCl_2 -35 m/o LiCl). The cooled salt phase was mechanically separated from the zinc and was then heated with magnesium sesquicarbide (Mg_2C_3) at 650°C for $\frac{1}{2}$ hr to form uranium monocarbide. Volatile components were then removed by heating to approximately 800°C under vacuum. X-ray diffraction analysis of the product indicated that UC is the major component and UO_2 a minor component. An analysis of the product for oxygen is being performed to determine the amount of UO_2 impurity.

c. Properties of Molten Chloride Systems. The systems LiCl-KCl-MgCl_2 and LiCl-NaCl-MgCl_2 are being considered as fluxes in liquid metal recovery processes for uranium-plutonium fuel. The flux compositions of greatest interest are those containing more than 50 m/o MgCl_2 and having a melting point below about 600°C. The liquidus temperatures in the MgCl_2 -rich portion of the LiCl-KCl-MgCl_2 and LiCl-NaCl-MgCl_2 systems have been determined for the purpose of finding a suitable flux composition. The range of flux compositions in the LiCl-KCl-MgCl_2 system which contains about 50 to 60 m/o MgCl_2 has been found to satisfy the two requirements. In the LiCl-NaCl-MgCl_2 system, the requirements are met in the flux composition range of 50-55 m/o MgCl_2 . In the LiCl-KCl-MgCl_2 system, a liquidus temperature as low as 350°C may exist in the area containing less than 40 m/o MgCl_2 and more than 30 m/o of both LiCl and KCl . In the LiCl-NaCl-MgCl_2 system, a minimum liquidus temperature of about 435°C has been indicated in preliminary thermal analysis studies.

d. Densities of Liquid Metals and Salts. A pycnometric technique for the determination of the densities of molten metals and molten salts has been developed. The technique has been used for the determination of the

densities of KCl at 783°C and of cadmium at 395°C. The densities were determined to be 1.523 g/cm³ and 7.92 g/cm³, respectively, as compared with literature values of 1.521 g/cm³ for KCl¹² at 782°C and 7.94₂ g/cm³ for cadmium¹³ at 395°C.

4. Calorimetry

A series of combustions of hafnium diboride in fluorine has been completed. A preliminary value of -76.6 kcal/mole for the heat of formation of a hafnium diboride sample of composition HfB_{2.02} was calculated from the heat of combustion of the sample and from auxiliary data.

¹²E. R. Van Artsdalen and I. S. Vaffe, J. Phys. Chem. 59, 118 (1955).

¹³H. T. Greenaway, J. Inst. Metals 74, 133 (1947-1948).

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Core Physics

It has been determined that the power coefficient can be represented by

$$\begin{aligned} d \ln k = & (2.6 + 0.19 T_R) 10^{-5} dT_R - 0.019 d \ln \rho_r \\ & - 1.9 \times 10^{-5} dT_c + 0.19 d \ln \rho_c - 0.37 \times 10^{-5} dT_M, \end{aligned}$$

(Doppler term omitted), where

T_R = Reflector coolant temperature, °F

T_c = Core coolant temperature, °F

T_M = Fuel temperature, °F

ρ_r = Reflector coolant density

ρ_c = Core coolant density.

The power coefficient, which is inherently negative, is also dependent on the rate of rise in power. For example, the faster the power increase, the more strongly negative is the power coefficient.

The coolant density terms can be related to temperature changes at constant pressure (no boiling) by

$$d \ln \rho = \frac{\partial \ln \rho}{\partial T} dT.$$

2. Fuel and Core Design

The first phase of the flow-stability verification tests with AARR fuel has been completed. The excessive vibrations and static deflections observed in the first tests were definitely induced by the large keys used to hold the fuel plates in place. Replacement of the keys with welded wire spacers has provided excellent hydraulic flow characteristics.

Flow tests were conducted at 100°F with stainless steel Type 304 dummy fuel test assemblies (FTS-4 and FTS-5) and with an aluminum Type 1100-H14 dummy fuel test assembly (FTA-5) to verify the stability of this design. The stainless steel assemblies experienced no fuel plate deflection at water velocities up to 65 fps. The aluminum assembly,

which approximates the elevated operating temperature strength conditions for stainless steel, experienced no fuel plate deflections at water velocities up to 50 fps.

Unfortunately, the experimental findings do not agree with the analytical models and the theoretical predictions obtained from published data. Effort is continuing on the development of an empirical relationship between plate instability and coolant velocity that will accurately describe the AARR test findings.

3. Beam Tubes for Experimental Facilities

The preliminary beam tube analysis for the AARR facility was completed by Franklin Institute early in March 1964. The analysis, consisting of thermal gradient and stress studies, was required to set the bounds for the allowable wall thicknesses for aluminum tubes. In addition, a two-dimensional electrical analog was constructed to determine the temperature distribution and cooling requirements.

These studies basically consisted of the following: 1) for a fixed inside diameter and heat-generation rate, the thermal gradient, average temperature, inside wall temperature, and cooling water temperatures were determined as a function of beam tube wall thickness; 2) for a fixed inside diameter, the total heat flux was integrated to determine total heat removal as a function of wall thickness; 3) from the information obtained above, combined stresses as a function of thickness were obtained for 4.25-in.- and 6.25-in.-ID tubes at 800-psig and 240-MW operation. The combined stress consists of the pressure stress, thermal stress, and discontinuity stress in the region of the hemisphere-cylinder junction.

The results, based on annular flow in a jacketed beam tube with a water inlet temperature of 175°F at the tube tip, indicate that outside cooling alone is sufficient to maintain metal temperatures within design limits. The maximum inside metal wall temperature was determined by computation and electrical analog for three different flow rates. The inside metal wall temperature can be kept below 300°F with reasonable flow rates. A portion of the results are summarized below for the largest beam tube (6.25-in.-ID with a 0.475-in. cylinder wall graduated to 0.2375 in. at the tip), in which the greatest stress occurs. Values are for a jacket channel width of 0.0625 in. and three different cooling rates. At 240-MW reactor operation, 800 psig pressure is required; this results in a compressive stress caused by external pressure of -4436 psi in all cases given below.

Case	Flow Speed (jacket), ft/sec	Flow (jacket), gpm	Max Metal Temperature, °F	Max Combined Compressive Stress (psi)
1	~6	~24.5	303	-9017
2	~12	~49.0	271	-8938
3	~24	~90.0	252	-9077

The maximum combined stress occurs at the junction of the hemispherical tip and the cylinder. The maximum pressure stress of -5722 psi occurs in the cylinder 2.77 in. away from the junction; at this point the combined stress is -8834 psi. The maximum thermal stress of -4943 psi occurs in the hemispherical tip 0.62 in. away from the junction; at this point the combined stress is approximately -8900 psi.

4. Critical Experiments

Fuel delivery for the AARR Critical Experiment is now scheduled on a firm basis. Enough bare uranium metal for the first loading is due in June and July. If no difficulties are encountered during fuel inspection or cladding application, loading of the first experimental core will begin in August.

Delivery of rolled hafnium plates for control blades is scheduled for May. The shopwork necessary to assemble control blades will not delay the startup. After trimming, machining to size, and assembly into composite control blades, the blades will be installed in the critical facility and operated for several cycles to test their mechanical integrity.

Study of flux-mapping schemes indicates that the accessible locations in the experimental core are insufficient to provide a satisfactory map. The total number of foil locations required for flux mapping is now estimated to be 116. Consequently, attention is being directed towards methods of removing one-third of the core for sample retrieval following foil-irradiation runs. Since some elements must be removed to retrieve foils, it may be advantageous to remove all the foils in this way in spite of the objection that fission product radiation will limit handling time severely.

An additional fast-fill interlock has been proposed to back up existing protection against pumping in moderator and reflector water at the fast rate at, or near, criticality. The proposed interlock would be activated by the recorder for channel I or II log count rate meter and would shut off the pumps at a subcritical level unless the operator (or the existing interlock with channel V log level instrument) closes the fast fill valve first.

B. Magnetohydrodynamics (MHD)

1. MHD Power Generation - Jet Pump Studies

Series operation of jet pumps did not show any significant improvement over the operation of a single unit. The difficulty is in properly producing velocity matching in these units. It appears, that combining the units requires a unique approach.

A new jet pump nozzle and combiner were designed to make such staging possible. In this design a compound nozzle, consisting of one central and two coaxially concentric nozzles connected to the vapor source, is used. The feed to the nozzles is orificed so that exhaust velocity is progressively increased in steps from the first outer concentric nozzle to the center unit. The nozzles are stepped to fit into the taper of the combiner, so that a stepwise acceleration of entering liquid without a great difference in velocities is provided. As the liquid enters the first nozzle, it is accelerated and subsequently injected into the region of the next higher velocity exhaust. The new units have been constructed and performance studies are progressing.

C. Regenerative EMF Cells

1. Sodium-Bismuth Bimetallic Cell

Preliminary engineering studies are being made of the sodium-bismuth bimetallic cell. With NaI-NaCl eutectic as the electrolyte, the apparent current densities at the anode were 500 mA/cm^2 at 0.35 V, whereas the apparent current densities at the cathode were 50 mA/cm^2 , because of the larger cathode area. When operating near the freezing point of the electrolyte, sodium utilization efficiency was as high as 86%.

Runs made with pure NaOH as the electrolyte at temperatures near its freezing point resulted in current densities and efficiencies that were similar to those obtained with the NaI-NaCl electrolyte. Irreversible sodium transfer increased significantly at temperatures somewhat above the freezing point of the electrolytes, thereby lowering the overall cell efficiency. Improvement of the operation of the cells might be possible by lowering the operating temperatures of the cells by using mixed halide electrolytes with lower melting points, or by substituting a mixed halide-hydroxide electrolyte for the pure NaOH electrolyte.

2. Corrosion Tests with Recrystallized Alumina

Static corrosion tests of recrystallized alumina, which is being considered for use as an electrical insulating material in bimetallic EMF cells were conducted. The alumina was immersed in liquid tin and liquid bismuth at temperatures as high as 1000°C . The duration of the tests varied from 24 to 76 hr. The alumina was found to be apparently compatible with bismuth and tin at the test temperatures.

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Fuel-Coolant Chemical Reactions

a. Reaction of Aluminum-Uranium Alloy and 6061 Aluminum Alloy with Steam. Studies of isothermal metal-water reactions are being undertaken in an effort to accumulate data that will allow a more direct comparison between results of laboratory experiments, data obtained in TREAT experiments, and results of reactor meltdowns.

Studies of the isothermal reaction of aluminum-17 w/o uranium alloy (SL-1 composition) with steam were completed during the month (see Progress Report for February 1964, ANL-6860, p. 97). The reactions were carried out over the temperature range from 1200 to 1600°C by the levitation method. In this method, a small spherical sample of the alloy is simultaneously heated and levitated by means of a radiofrequency field while steam is flowed over the sample. The results of the experiments showed that the reaction is described by the same cubic rate law that describes the reaction of aluminum with steam at temperatures below 1300°C.¹⁴

A short series of isothermal experiments has been started for the reaction of 6061 aluminum with steam. The 6061 aluminum contains 1.0% magnesium, 0.6% silicon, 0.25% copper, and 0.25% chromium. The determination of the possible effects of the alloying agents on the reaction is pertinent to the program directed toward achieving correlations between variously derived data, since the 6061 aluminum alloy was the base alloy used in the SPERT-ID core. Preliminary results of studies carried out by means of the levitation method in the temperature range from 1300 to 1600°C indicate that the presence of the alloying elements has little effect on the aluminum-steam reaction.

b. Laser-beam Heating Applications. It has been previously reported (Progress Report for February 1964, ANL-6860, p. 97) that when particles of zirconium (or aluminum) are heated by means of a laser beam while submerged in water, a steam-hydrogen bubble is generated which, under certain conditions, collapses after a period of about 10 msec. As a result of this phenomenon, water contained in an open tube is driven violently against the bottom of the tube, creating a so-called "water hammer." High-speed motion pictures have been taken of the bubble formation and are being used to determine volume-time relationships. Simple geometric shapes, such as hemispheres, cylinders, and spheroids, are being assumed for the calculations of the volume of the bubbles formed. A mathematical model is being developed which will allow quantitative comparisons to be made of bubble formation and collapse under various conditions.

¹⁴Chemical Engineering Division Summary Report; October, November, December 1962, ANL-6648, p. 198

c. Studies in TREAT of Oxide-core, Metal-clad Fuel Pins. Three additional experiments were performed in TREAT to obtain metal-water reaction data during very energetic reactor transients. The tests were carried out with fuel pins of three different compositions. A single fuel pin submerged in water at room temperature was used in each test. The conditions and results of the test are summarized in the following:

	CEN Transient Number		
<u>Reactor Characteristics</u>	<u>172</u>	<u>173</u>	<u>174</u>
Integrated Power, MW-sec	438	867	860
Reactor Period, msec	63	39	39
Energy Input, cal/g	394	694	688
<u>Fuel-pin Characteristics</u>			
Core Material	93.5 w/o U, 5.0 w/o Zr, 1.5 w/o Nb	UO ₂	UO ₂
Cladding	none	Zr-2	SS-304
Pre-irradiation	1.5% burnup U ²³⁵	none	none
Enrichment, % U ²³⁵	20	11.2	11.2
<u>Results</u>			
% Metal Reacted with Water	51.3	79.8	35.5
Final Geometry of Fuel Specimen	fine particles	fine particles	globules and particles
Peak Pressure, psi	-	480	111
Maximum Pressure Rate, psi/msec	-	1.5	1.6

The results of these tests and of previous tests (see Progress Report for December 1963, ANL-6810, p. 53, and Progress Report for February 1964, ANL-6860, p. 98) indicate that pre-irradiation, at least up to 1.5% burnup of U²³⁵, does not influence the extent of the uranium-water reaction.

The results from the present experiments and the accumulated data from previous TREAT experiments with oxide-core fuel pins indicate that (1) the extent of the reaction of stainless steel cladding is a continuous function of transient energy input in the energy zone in which core and cladding is destroyed (above about 200 cal/g of oxide core), and (2) the extent of reaction of Zircaloy-2 cladding with water increases linearly with energy input to about 800 cal/g of oxide core, at which point the extent of reaction shows a very sharp increase.

2. Metal Oxidation-Ignition Studies

a. Thermal Conductivity of Metal Powders. The effect of particle size, temperature, pressure, and degree of oxidation on the thermal conductivity of metal powders is being studied to gain a better understanding of the mechanism of ignition of metal powders, in order that effective preventive measures against accidental ignitions may be formulated.

The effect of mesh size and degree of oxidation on the thermal conductivity of zirconium powder has been studied. In one series of experiments, the thermal conductivities of -70 +80 mesh zirconium powder at various stages of oxidation up to 86.5% zirconium oxidized were measured in nitrogen at 24°C. The thermal conductivity values measured at atmospheric pressure decreased with increasing oxide content. The values [in cal/(cm)(sec)(°C)] were 7.4×10^{-4} for unoxidized zirconium, 7.0×10^{-4} for 13.3% zirconium oxidized, and 4.4×10^{-4} for 86.5% zirconium oxidized.

Thermal conductivity measurements with -230 +325 mesh zirconium powder, oxidized to various degrees up to 15.5% zirconium reacted, were carried out under the same conditions that were employed in the study of the -70 +80 mesh powders. The trend in conductivities with extent of oxidation was the same as that observed with the coarser zirconium powder.

b. Aluminum- U_3O_8 Reaction. Additional studies were made of the aluminum- U_3O_8 thermite reaction (see Progress Report for February 1964, ANL-6860, p. 100). The report¹⁵ of a very violent thermite reaction in aluminum- U_3O_8 cermet led to the re-examination of this reaction.

Earlier studies of the reaction employed cold-pressed compacts made from mixtures of coarse U_3O_8 powder (average particle size, 109μ) and fine aluminum powder (26μ) or from mixtures of fine U_3O_8 powder (12μ) and fine aluminum powder. In the present tests, the coarse U_3O_8 powder was compacted with coarse aluminum powder (109μ). The compacts were rapidly inserted into a furnace preheated to 1250°C. The reaction occurring between the U_3O_8 and aluminum was incomplete and less vigorous than previous reactions in which the 109μ aluminum powder was used.

B. Fast Reactor Safety Studies

1. Niobium-clad Uranium Carbide Fuel Pins

Interest in using ceramic fuels in high-temperature, high-power-density, liquid-metal-cooled reactors prompted the transient studies on

¹⁵J. D. Fleming and J. W. Johnson, *Nucleonics* 21(5), 84 (1963).

UO₂ and uranium sulfide in the TREAT reactor. Uranium carbide clad with Nb-1 w/o Zr has been studied in meltdown experiments. The uranium carbide fuel was prepared in short cylindrical pellets, 0.648 cm in diameter, and stacked to a length of approximately 27.9 cm in an evacuated niobium tube of 0.030-cm wall thickness. Two samples were given destructive power bursts inside the transparent meltdown facility to obtain a photographic record of the sample behavior.

Both specimens failed during TREAT bursts of approximately 30 MW-sec. Maximum cladding temperature recorded prior to failure was about 500°C. The estimated maximum cladding temperature was 900°C, from which it was deduced that the maximum central fuel temperature was about 1200°C, and the maximum fuel temperature about 1500°C. (The maximum fuel temperature does not occur at the center during transient TREAT tests, because of the thermal neutron flux depression inside the fuel.)

Postmortem examination of the test samples revealed that both had failed just below the sample vertical midplane. The niobium cladding had burst, forming a gaping tear. One specimen (see Figures 24 and 25) still retained closely packed fragments of shattered uranium carbide pellets in the failure region. Finely divided uranium carbide was intermixed with the larger fragments, and was found deposited on the mirrors and windows of both capsules. The uranium carbide pellets showed no signs of reaching melting temperatures, and the broken surfaces of the fragments indicated simple brittle fractures.

Xrays of typical fuel elements showed spacings between the upper end of the pellet stack and the internal restrainer ranging between 0.23 cm and 0.48 cm, as contrasted with the reference spacing of 0.554 cm. Calculations of radial and axial thermal expansions for the two destructive transients indicate that extreme axial loading of the uranium carbide pellets could have occurred.

The centralized fragmentation of the uranium carbide fuel pellets, the appearance of the cladding breach, and the condition of the broken surfaces indicate that failure of the samples occurred as a result of extreme internal pressures which were mechanical rather than hydrodynamical in nature. It can be surmised that the cladding split under unusual circumferential stresses caused by a combination of compressive forces resulting from the thermal expansion of the higher-temperature fuel pellet stack against the elastic restraint of the cooler niobium container. Under these conditions, the elastic restoring forces of the cladding tube would cause the ejection of the fuel fragments upon the restraint relief afforded by the yielding of the tube wall.

Figure 24. Appearance of Failure Region for Niobium-clad UC Sample

Figure 25. Appearance of Cracked UC Pellets from the Region of Failure

2. Meltdown Experiments of Pre-irradiated Samples

Transient experiments are being performed in TREAT to study the effects of fission products on fuel failure and movement. A group of twenty 6% enriched EBR-II Mark-I pins have been encapsulated for steady-state irradiation in Materials Testing Reactor. The twenty specimens, which have been shipped to Idaho, are specified for nominal burnups in the range from 1.5 to 3.0 a/o.

3. Estimates of Pressure Pulse for Sodium Loop Experiments

Calculations of the transient temperature by means of the ARGUS computer code have been used to prepare input for the pressure pulse code (see Progress Report for December 1963, ANL-6810, p. 53) to obtain estimates of maximum loop pressures under severe but near-normal and "accident" conditions. Power input to the coolant at initiation of boiling was estimated from the temperature calculations and used in the pressure computations.

Two power excursions were studied:

- (1) $k_{ex}(0) = 1.56\%$, sample pin energy input during the power peak of 122 cal/gm of fuel (or 3000 Btu for a 91-pin cluster). Based on experimental results with stagnant sodium, this excursion should cause extensive pin failure.
- (2) $k_{ex}(0) = 1.95\%$, sample pin energy input during the power peak of 244 cal/gm of fuel (or 6000 Btu for a 91-pin cluster).

For the integral sodium loop, which can accommodate 1-7 fuel pins, two cases were calculated: a single pin surrounded by six dummy pins (the arrangement of current interest), and seven heated pins. In the first case, maximum pressure rises of 0.3 atm and 2.4 atm were calculated for excursions (1) and (2), respectively. Maximum pressure rises were higher for seven heated pins; 1.5 atm and 6.5 atm were obtained for the two excursions.

Estimates for the large loop were larger, principally because of the larger quantity of sodium downstream from the test pins, which must be accelerated by the vaporizing sodium. For the test section with surge tank, maximum pressure rises of 8.5 atm and 30 atm were estimated. Removal of the surge tank led to calculated pressure rises of 29 and 102 atm, respectively.

4. Small TREAT Loop

During the mockup study of the in-cave remote-handling equipment, a number of interferences were discovered between portions of the equipment

The area has been modified to correct the faults, and the equipment is being readied for final demonstration. An assembled loop is being charged with sodium and prepared for the demonstration.

5. Large TREAT Loop

a. Flowmeters. The flowmeters for 2- and 3-in. pipe used in the large TREAT loop have been calibrated in the temperature range from 400 to 800°F. The calibration is essentially linear at flow rates up to 60 gpm, a result that lends confidence that extrapolation to the loop maximum flowrate of 160 gpm is valid. A zero voltage reading is not obtained at no flow, since a slight reversing potential is measured at that time. However, this does not detract from the effectiveness of the calibration. The temperature coefficient of the flowmeter is very small, so that significant error will not be introduced under varying temperature conditions. The average coefficient between 400 and 760°F is equal to 0.0005 mV/°F which is equivalent to a flowrate of 0.01 gal/min.

b. Construction. The cold trap and associated heat exchanger are 65 and 50% completed, respectively. In addition, the plugging indicator is approximately 90% complete.

Mass-spectrometer testing of the balls and seats of the P-K 4-in. valves has been completed. Seating satisfactorily meets the requirements of the specifications. The next step will be to test the closure welds at the base of the bellows. If the results of these leak tests are up to expectation the valves will be crated and shipped to Idaho.

A study of the steps for unloading sodium from the test section were reviewed. This study also included the procedure for preparing this test section for shipment back to Lemont after an experiment has been completed.

VI. PUBLICATIONS

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STUDIES OF LITHIUM HYDRIDE SYSTEMS. I. SOLID-LIQUID EQUILIBRIUM IN THE LiCl-LiH SYSTEM

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