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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

October 1962

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Issued November 15, 1962

Operated by The University of Chicago
under
Contract W-31-109-eng-38
with the
U. S. Atomic Energy Commission

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FOREWORD

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

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

The last six reports issued
in this series are:

April 1962	ANL-6565
May 1962	ANL-6573
June 1962	ANL-6580
July 1962	ANL-6597
August 1962	ANL-6610
September 1962	ANL-6619

TABLE OF CONTENTS

	<u>Page</u>
I. Water-cooled Reactors	1
A. EBWR	1
1. Further Destripping of Spikes to Gain Reactivity for Operations above 60 Mw	1
2. Operation of the Reactor above 60 Mw	2
3. EBWR Transfer Functions	2
B. BORAX-V	3
1. Operations and Experiments	3
2. Modification and Maintenance	4
3. Analysis	5
4. Procurement and Fabrication	5
5. Development and Testing	6
II. Liquid Metal-cooled Reactors	8
A. General Research and Development	8
1. ZPR-III	8
2. Preparations for ZPR-VI and ZPR-IX	13
3. AFSR Experiments	14
B. EBR-I	15
C. EBR-II	15
1. Reactor Plant	15
2. Power Plant	18
3. Sodium Boiler Plant	18
4. Fuel-Cycle Facility	18
5. Fuel Development	20
6. Process Development	22
7. Training Program	25
D. FARET	25
III. General Reactor Technology	28
A. Applied Reactor Physics	28
1. Measurements of Fast-neutron Spectrum	28
2. High-conversion Critical Experiment: ZPR-VII	28
3. Absolute Measurement of ν for U^{235} and Cf^{252}	29

TABLE OF CONTENTS

	<u>Page</u>
4. Elastic Scattering of Neutrons	30
5. Spontaneous Fission Spectrum of Cf ²⁴⁸	33
6. Theoretical Physics	34
B. Reactor Fuels and Materials Development	36
1. Corrosion Studies	36
2. Ceramic Fuels	37
3. Nondestructive Testing	39
C. Heat Engineering	41
1. Heat Transfer between Fluids of Low Prandtl Number in Double-pipe Heat Exchangers	41
2. Studies of Boiling Liquid Metals	43
D. Chemical Separations	43
1. Fluidization and Volatility Separations Processes	43
2. Chemical-Metallurgical Process Studies	48
IV. Advanced Systems Research and Development	51
A. Argonne Advanced Research Reactor (AARR)	51
1. Site and Buildings	51
2. Research and Development	52
B. Undersea Application of Nuclear Power	54
V. Nuclear Safety	55
A. Thermal Reactor Safety Studies	55
1. Metal Oxidation and Ignition Studies	55
2. Metal-Water Studies	56
B. Fast Reactor Safety Studies	56
1. Meltdown Experiments	56
2. Integral Sodium Loop Development for Experimental Meltdown Studies	58
3. Operation and Maintenance of TREAT	59
VI. Publications	60

I. WATER-COOLED REACTORS

A. EBWR

Additions and modifications to the EBWR plant have been completed which permit heat dissipation at power levels up to 100 Mwt. The plant has been authorized to operate at its full capacity and operational testing at approximately 85 Mwt level has been reached.

The composition of the current core fuel loading (Core 1A) consists of the following:

<u>Subassembly Type</u>	<u>Number</u>
Spike (Rod Type Elements)	32
Natural Uranium (Thick Plate)	8
Natural Uranium (Thin Plate)	1
1.44% Enriched (Thick Plate)	52
1.44% Enriched (Thin Plate)	54
Total Number of Subassemblies	<u>147</u>

1. Further Destripping of Spikes to Gain Reactivity for Operations above 60 Mw

Sixteen boron strips were removed from the spike fuel elements to provide the additional reactivity required to increase the power of the reactor above the 60 Mwt reached last month. The removal of the strips left sufficient control in the reactor to meet the shutdown requirements noted in the EBWR Hazards Report.

The ring of 28 spikes now contains:

20 spikes having 1 strip/spike
 4 spikes having 2 strips/spike
 4 spikes having 0 strip/spike.

There is an average of one boron strip per spike in the ring, with four strip-less spikes. The gain in reactivity was $\sim 2.1\% \Delta k/k$. The other four spikes in the reactor are located on the periphery of the core.

The boron strips appeared in good condition. An examination of the strips did not reveal any surface cracking and were ductile (i.e., bowed to arcs readily). The fuel rods also appeared to be in good condition. There was no indication of bowing and the oxide coating on the surface was uniform.

2. Operation of the Reactor above 60 Mw

The introduction of 2.1% reactivity was expected to permit the power to reach a level of 80 to 85 Mwt with all of the boric acid removed from the water. The reactor, however, achieved a level of ~85 Mwt (with near equilibrium xenon) with excess reactivity to spare, the amount being equivalent to that represented by 1 gm/gal of boric acid.

It appears, therefore, that there has been an improvement in the power distribution and hydrodynamic flow in the core. Power runs at 65 Mw and higher will be made to determine the causes for the change in power distribution and the changed hydrodynamic behavior in the coolant.

Activity levels have increased considerably in the subreactor room nozzles, feedwater filters, and the ion-exchange system. This indicates that some descaling is occurring from the original plate-type fuel elements during the higher power runs. The scale deposits resulted from deposition of corrosion products arising from the use of dummy aluminum fuel boxes which have since been removed from the reactor.

3. EBWR Transfer Functions

The transfer functions of EBWR were measured at the following conditions:

- (1) 65 Mw, 590 psig, with a boric acid concentration of 2.35 gm/gal. Control rods 1-8 at 48 in. and center rod at 30 in.
- (2) 75 Mw, 590 psig, with a boric acid concentration of 1.1 gm/gal. Control rods 1 and 2 at 41 in., 3 through 8 at 40.5 in., and center rod at 30.0 in.

The measurements for the 65-Mw test were made over a frequency range from 0.89 to 8.9 cps, and for the 75-Mw test over a range from 0.067 to 8.9 cps. Data were recorded with the analog sine-cosine cross-correlator and digital data system. Analog recordings were made of noise and transient response. Root-mean-square (rms) noise was measured with an analog computer. The autocorrelation function and power-density spectrum were calculated from the digital noise recordings.

Analog and digital recordings of reactor noise were made under the following conditions:

- (1) 78 Mw, 590 psig, control rods 1 through 8 at 36.5 in., center rod at 30 in. Boric acid concentration 1.1 gm/gal.

- (2) 75 Mw, 590 psig, control rods 1 through 8 at 34.7 in. and center rod at 39.75 in. Boric acid concentration 1.0 gm/gal.
- (3) 85 Mw, 590 psig, control rods 1 through 8 at 35.2 and center rod at 39.75 in. Boric acid concentration at 1.06 gm/gal.
- (4) Same conditions Item (3), except a high-pass filter was used to eliminate the low-frequency noise below 2.5 rad/sec.

Recordings were also made of reactor pressure, fuel element inlet and outlet flow, and subcooling temperature.

The stability extrapolation based on the 75-Mw transfer function data indicated stable operation to greater than 100 Mw of power output, an increase over the 85 Mw predicted from the 60-Mw transfer function data.

The power distribution in the core as established by control rod positions has an effect on reactor flux noise. The rms values of noise measured for conditions 1, 2, and 3 were 7.5%, 4.7%, and 5.1%, respectively. The 2.8% difference between conditions 1 and 2 occurred for frequencies less than 0.065 cps.

B. BORAX-V

1. Operations and Experiments

Procedures for replacing the aluminum rivets by nickel rivets in the boiling fuel assembly boxes required unloading all the fuel from the reactor. This was done safely by dissolving 20 gm/gal of boric acid in the reactor water to maintain the reactor subcritical, even if all the control rods had been removed. After repair and reloading, six flux-wire and five miniature ion-chamber pressure-tight thimbles and a fixed, miniature ion chamber were inserted in the core, as was the rotating oscillator rod and fuel assembly. A magnetic, drag disc-type downcomer flowmeter was attached to the outside of the core structure shroud, and a strain-gauge-type pressure transducer for measuring transient pressure was located just above the core. Installation of the thimbles and oscillator rod reduced boiling core excess reactivity by about 1%.

Zero-power experiments at conditions up to 600 psig, 489°F, are being run. Control rod calibrations, with boric acid in the reactor water to permit almost complete control rod withdrawal, indicate that the differential reactivity worth of the 9-rod bank at 489°F is about $\frac{2}{3}$ to $\frac{1}{2}$ the value of the measurement made previously at room temperature with the present core. The differential worth at room temperature is about 80% of the value measured earlier with the original set of control rods in

the core with 36 poison rods. The reactivity worth of boric acid in the reactor water at 489°F is about 0.2% $\Delta k/k$ per gm per gal. This compares with a value of about 0.6% at room temperature measured in the same core. The average temperature effect measured from 119° to 489°F, corrected for boric acid, is -0.013%, $\Delta k/k/^\circ\text{F}$. Experiments to measure the temperature effect without boric acid in the reactor water have started.

Equipment for determination of transfer functions with an analog computer and null-balance systems has been set up in the reactor building and is being checked out. The drive for the rotating oscillator rod has been installed and is operative.

Experimental regenerations of the make-up water demineralizer, using three hot (110°F), 10% caustic solution 24-hr soaks to remove silica from the anion resin, have restored capacity of the ion exchange to three-fourths of rated value, in contrast with a value of one-third before this. Further treatments are expected to continue to increase this value.

A piping change was made to permit transfer of water from the electric preheat system to the bottom of the reactor vessel, thereby making it possible to maintain the temperature of the bottom head more nearly equal to the temperature of the remainder of the vessel. Satisfactory temperature distribution in the lower head is now obtained.

2. Modification and Maintenance

Modification of all 67 boiling fuel assembly boxes at the Idaho site was accomplished. The twelve $\frac{1}{8}$ -in.-dia aluminum rivets attaching each stainless steel upper and lower grid to the aluminum boiling fuel assembly boxes were replaced with twenty $\frac{3}{16}$ -in.-dia "A"-nickel rivets. One box was filled with dummy stainless steel fuel rods and is being autoclave-tested at conditions of 600 psig and 489°F.

The rivets in instrumented boiling fuel assembly I-2 were replaced after removal of the inlet flowmeter. Upon attempting to recalibrate the flowmeters in the air-water test loop, it was found that the pickup coils of both meters had become inoperative because of low insulation resistance resulting from leaks. Both coils, with leads, are being replaced by the vendor. They must be reinstalled on the flowmeters before the instrumented assembly can be returned to the reactor.

A brazed-on backup sleeve is now being designed for use on all flowmeter coils to eliminate the present Inconel-to-stainless steel weld where possible or to cover the weld on coils already installed.

The control rod seals were changed last month by replacing the Delrin guide bushing wipers with Teflon guide bushings; also, three control rod extension shafts were replaced with new shafts, and the remainder were ground to remove scratches resulting from the failure of the seal guide bushings. This has eliminated the difficulties with failure of these guide bushings because of high temperature on the control rod seals. Better stability and control-rod-seal cooling water flow has been obtained by damping the differential-pressure controller oscillations. Control rod drive performance has been satisfactory since these modifications were completed.

Some of the nuclear instruments were modified to improve their stability. Period meter amplifiers were relocated from the control room to the reactor building instrument room, and the linear neutron-flux amplifier was rebuilt to change the range switching system. The results are improved operation of the period and linear flux instruments.

3. Analysis

The theoretical dynamic model of the BORAX V reactor has been reduced to a form in which the $\Delta n/n_0 \Delta k_{ex}$ transfer function can be readily calculated with the aid of either of two IBM-704 digital codes. They are (1) the "BUM" Transfer Function Code (RE-132), ANL, and (2) the Block Diagram Reduction Code, Allis-Chalmers. The first set of problems correctly being programmed calculates the theoretical power transfer function for the natural-convection boiling core at a nominal power level of 20 Mw. Parameters to be varied in this first study are the void coefficient of reactivity, Doppler coefficient, and the fraction of total water mass in the reactor vessel at saturation temperature and pressure.

A simplified model has been derived for determining approximate optimized controller settings (as a function of reactor power level) for the steam pressure-control system.

4. Procurement and Fabrication.

a. Superheater Fuel - Replacements for the production fuel plates which had been previously rejected have been received from the contractor, inspected, and found to be satisfactory.

A total of 80 peripheral-type subassemblies have been brazed to date. Completion of 17 required elements of this type will require 85 subassemblies. Since two of the first 80 subassemblies brazed were damaged during subsequent machining operations, seven more subassemblies are required.

Welding assembly has been completed on all 13 standard central superheater fuel assemblies. Some bowing, measured in the 12-ft assembly length after welding, was straightened by localized heating.

Brazing of central instrumented subassemblies is now in progress. Of the nine required, five have been completed. No damage to thermocouples occurred during the brazing process. The first seven central-type elements were completed by heliarc welding risers and nozzles to ends of subassemblies. The elements are being checked for dimensional fidelity and bowing, and straightened as necessary.

5. Development and Testing

a. Corrosion of Type 304 Stainless Steel in Superheated Steam - Various surface preparations on Type 304 stainless steel specimens were exposed to steam at 650°C and 600 psi containing about 0.2 ppm oxygen for a period of one week. The order of increasing corrosion for treatments of the surfaces were (1) machined, (2) grit blasted, (3) wet ground, (4) etched, and (5) electropolished. Tests with heavily cold-reduced Type 304 stainless steel (of predominantly ferritic structure) and annealing procedures suggest that a thin layer of transformed ferrite produced on the surface of the austenitic stainless steel by cold work may be responsible for the improved resistance of the worked surfaces as compared with that of the electropolished surface.

With our refreshed autoclave system it was impossible to maintain stoichiometric amounts of oxygen and hydrogen in the steam at the 30-ppm oxygen level. Recombination at the test temperature reduced the output oxygen content to 1-3 ppm.

b. Advanced Superheater Fuel - AISI Type 406 stainless steel is being considered for use with superheat fuel elements in BORAX-V. Work has started on the development of improved techniques for hot roll-bonding of cermet fuel plates, with Type 406 stainless steel as cladding and matrix. Sample sandwiches of Type 406 stainless steel were seal-welded along the edges and heated and hot-rolled in air. The material was first descaled, and the two pieces for one compact were plated with 0.0002-in.-thick nickel. All edges of both compacts were then sealed by welding. No evidence of inclusions was found at the interfaces of the rolled plates. The two pieces which were only descaled prior to welding seemed to be bonded more thoroughly than the nickel-plated pieces. Material has been ordered so that full-sized fuel plates of the BORAX-V type can be similarly fabricated and examined for inclusions at clad-to-clad interfaces.

Preparations are being made to fabricate full-size experimental fuel plates from UO₂-Type 406 stainless steel by seal-welding an evacuated cladding-compact sandwich, heating and hot-rolling in air, and then descaling.

Brazing studies conducted on small plate samples of AISI Type 406 stainless steel have shown that either Coast Metals 60 or Permabraz 128 wets the material and flows satisfactorily if surface scale formation is prevented. Specimens which were descaled, plated with 0.0002-in.-thick nickel, and brazed with these two alloys respectively, have been submitted for corrosion testing in degassed and oxygenated steam.

c. Experimental Components - The magnetic drag disc down-comer flowmeter was assembled and calibrated, with the use of weights to simulate water flow at 489°F. With a dual-range switch on the read-out chassis, this meter should allow measurements over ranges from 0.25 to 2.5 and from 0.1 to 1.25 ft/sec with accuracies of 2 to 3% of full scale.

The salvage operation on instrumented boiling fuel assembly I-1 is nearly complete. Backup sleeves must be installed on the flowmeter coils and aluminum rivets must be replaced.

A study has been initiated to investigate dispersion strengthening of Type 406 stainless steel.

II. LIQUID METAL-COOLED REACTORS

A. General Research and Development

1. ZPR-III

Experiments were carried out in the program of studies with Assembly 41, a dilute metal system with 20% enriched uranium in the core. This assembly, with a critical mass of 490 kg U^{235} in a 439-liter core, is described in detail in ANL-6619 (Reactor Development Program Progress Report, September, 1962).

Among experiments performed were those to give measurements of reactivity coefficients and fission ratios, blanket and reflector experiments, and a mockup of a rotary control rod, containing boron carbide, in the radial blanket.

a. Central Fission Ratios - Ratios of fission rates of the various fissile isotopes were obtained at the core center with the use of absolute fission chambers of a basic design described by Kirn.¹ Measurements of counting ratios were made with the original Kirn counters on hand and also with gas-flow counters, with the same basic design but thinner chamber walls, and with interchangeable foils of fissile platings. Results of the measurements are given in Table I.

Table I. Central Fission Ratios in Assembly 41

Counters Used		Ratio Obtained	Value
Half No. 1	Half No. 2		
Natural U foil #2 in thin-wall gas-flow counter	Enriched U foil #3 in thin-wall gas-flow counter	$\sigma_f U^{238} / \sigma_f U^{235}$	0.0392
Kirn Counter #2 (Depleted U)	Kirn Counter #5 (Enriched U)	$\sigma_f U^{238} / \sigma_f U^{235}$	0.0366
U^{236} foils #27 and 28 in thin-wall gas-flow counters	Enriched U foil #3 in thin-wall gas-flow counter	$\sigma_f U^{236} / \sigma_f U^{235}$	0.0902*
Kirn Counter #24 (U^{236})	Kirn Counter #5 (Enriched U)	$\sigma_f U^{236} / \sigma_f U^{235}$	0.1103
Kirn Counter #18 (U^{233})	Kirn Counter #5 (Enriched U)	$\sigma_f U^{233} / \sigma_f U^{235}$	1.430
Kirn Counter #11 (U^{234})	Kirn Counter #5 (Enriched U)	$\sigma_f U^{234} / \sigma_f U^{235}$	0.286
Kirn Counter #21 (Plutonium)	Kirn Counter #5 (Enriched U)	$\sigma_f Pu^{239} / \sigma_f U^{235}$	1.164

*Tentative value.

¹Kirn, F. S., An Absolute Fission Counter, ANS Second Winter Meeting (1957).

The count rates of some newly plated foils of enriched and natural uranium were compared in the gas-flow counters with older foils. Agreement of count rates per gram of plating was good. However, recently prepared foils with U^{233} platings showed count rates in the flow counters about double those expected on the basis of the U^{233}/U^{235} fission ratio measured with the Kirn counters. The source of the anomaly is being sought. New platings of U^{236} material were tested also and results indicated some discrepancy. The U^{236}/U^{235} fission ratio obtained with the thin-walled gas-flow counters, containing the new U^{236} foils, is seen in Table I to be lower than the corresponding ratio obtained with Kirn counters. On the basis of previous experiments in other assemblies, the heavier-walled Kirn chambers should give lower fission rates for the threshold isotopes.

b. Sample-size Effects in Measurements of Central Reactivity Coefficient - A series of worth measurements at the core center were made with samples of materials varying in size and neutron capture or scattering properties. The materials used and sample dimensions are given in Table II together with the measured worths per kilogram. In

Table II. Measurements of Central Reactivity Coefficient - Sample-size Effects

Material	Sample Space (in.)	Volume % Material	Sample Worth ^(a) (in hours)	Reactivity Coefficient ^(a) (in hours/kg)
Steel-clad Plates of Enriched B Mixture (94 w/o B, B = 92 a/o B ¹⁰)	$\frac{1}{2} \times 2 \times 2$	21	-22	-1460
	$2 \times 2 \times 2$	5.2	-23	-1450
	$2 \times 2 \times 2$	12.7	-29.5	-1088
Tantalum	$\frac{1}{2} \times 2 \times 2$	100	-12.1	-23.0
	$2 \times 2 \times 2$	25	-12.8	-24.3
	$2 \times 2 \times 2$	48	-23.7	-23.6
	$2 \times 2 \times 2$	100	-44.4	-21.1
Thorium	$\frac{1}{2} \times 2 \times 2$	100	-4.50	-12.1
	$2 \times 2 \times 2$	100	-17.8	-11.8
Enriched B ₄ C (B = 94 a/o B ¹⁰)	$\frac{1}{2} \times 2 \times 2$	100	-62	-961
	$2 \times 2 \times 2$	100 in a $\frac{3}{4} \times 2 \times 2$ piece	-66	-684
Depleted Uranium	$\frac{1}{2} \times 2 \times 2$	100	-2.94	-5.3
	$2 \times 2 \times 2$	100	-12.3	-5.1
Polyethylene (CH ₂)	$\frac{1}{2} \times 2 \times 2$	100	23.8	+817
	$2 \times 2 \times 2$	25	22.2	+731
	$2 \times 2 \times 2$	50	50.7	+845
	$2 \times 2 \times 2$	75	78.6	+879
	$2 \times 2 \times 2$	100	99.5	+838

(a) Values reported here have been corrected for canning and spacing materials.

those cases in which the material measured occupied less than the volume of sample space, the remainder of the space was filled with empty steel cans, perforated aluminum, or was left void. The particular procedure in each case was adapted to the sample geometry. The worth of steel or aluminum in each case and the values reported in the table are corrected to represent only the material being measured. The results indicate that capture self-shielding effects are prominent for B^{10} and are also present to a lesser degree for tantalum. With the polyethylene samples, the principal concern is with the effects of fuel proximity.

c. Measurements of Reactivity Coefficient at Core Radial Edge - Measurements of reactivity worths for materials at the core radial edge near the midplane were made by substitution of materials into sample spaces constructed at the front of drawers 1-J-12 and 2-V-20. For the fissile samples and boron materials, each sample space was $2 \times 2 \times \frac{1}{4}$ in., but $2 \times 2 \times 1$ -in. regions were used with the other materials. Table III presents measured sample worths and the reactivity coefficient in

Table III. Reactivity Coefficients at Core Radial Edge Near Midplane

Material	Sample Worth ^(a) (in hours)	Reactivity Coefficient (in hours/gm-mole)
Enriched Uranium (93.2%)	7.6	6.19
Depleted Uranium (0.2%)	2.0	0.198
Plutonium (94.5% 239)(c)	8.1	10.40
Enriched B_4C (b)	-12.9	-1.044
Aluminum	2.6	0.200
Al_2O_3	3.0	0.856
Arsenic(c)	0.27	0.053
Beryllium	3.6	0.136
Bismuth	3.0	0.491
Carbon	2.8	0.171
Polyethylene (CH_2)	8.0	1.75
Iron	2.7	0.146
Molybdenum	2.1	0.156
Sodium(c)	1.4	0.214
Niobium(c)	1.17	0.222
Nickel	3.5	0.180
Lead	3.45	0.485
Sulfur(c)	0.34	0.172
Steel (304)	3.0	0.165
Tungsten(c)	0.58	0.027

(a) Two $\frac{1}{4} \times 2 \times 2$ -in. sample spaces for enriched uranium, plutonium, and enriched B_4C measurements; two $1 \times 2 \times 2$ -in. samples for all other materials.

(b) Boron = 90.7 a/o B^{10} .

(c) Samples clad in stainless steel; sample worths quoted have been corrected for steel effects.

inhours/gm-mole. Although these experiments for local reactivity coefficient indicated that aluminum would be worth more than depleted uranium, later experiments (see Section f) showed that the aluminum was worth less than depleted uranium when substituted throughout a large volume of the blanket.

d. Heterogeneity Experiments - A measurement of the heterogeneity effect of the arrangement of core fuel, with $\frac{1}{8}$ -in.-thick enriched uranium plates, was made to allow comparison of experimental critical mass with theoretical calculations for a homogeneous system. In one experiment, thirty-eight $\frac{1}{8}$ -in. columns of enriched uranium contained in two wedge-shaped sectors of Half No. 1 were "bunched" into nineteen $\frac{1}{4}$ -in. columns. A gain of reactivity of 52.1 inhours was obtained. A total of 44.25 kg enriched uranium was involved; if the results are extrapolated to the enriched uranium content of 520 kg for a just-critical core (490.3 kg U^{235}), an increase of 612 inhours would be obtained.

Another experiment was performed in which the fuel was unbunched. First, twenty-three $\frac{1}{8}$ -in. columns of enriched uranium in a wedge sector of Half No. 1 were reconstructed with double $\frac{1}{16}$ -in. columns. These $\frac{1}{16}$ -in. columns were then separated, or unbunched, giving an 8.6-inhour loss in reactivity. A total of 26.7 kg enriched uranium was unbunched, and, in again extrapolating to 520 kg, a 167-inhour loss was obtained.

From the bunching and unbunching results, the 490-kg U^{235} assembly core, with its $\frac{1}{8}$ -in. fuel plate arrangement, is estimated to be 335 inhours more reactive than would a homogeneous core with the same average composition. This is equivalent to about 0.8% $\Delta k/k$.

e. Rotary Control Rod with B_4C - A means of reactivity control in fast power reactors has been proposed whereby a rotary control rod containing eccentrically loaded enriched B_4C would be placed in the radial blanket just outside the core. An experiment was performed to evaluate such a system with a rod of about 6-in. diameter. Worths of depleted uranium and enriched boron carbide were measured in a column $2 \times \frac{1}{2}$ -in. in cross section by 16 in. in axial length. The column was first placed in a depleted uranium radial blanket drawer 1-P-8, centered $\frac{1}{2}$ in. away from the radial core edge. Next, the column position was shifted to drawer 1-P-6, centered 6 in. from the core edge. Table IV presents the measured worths. The data indicate that a full core-length rod containing 30 gm B^{10} per inch would provide a reactivity variation of about 105 inhours from maximum to minimum worth.

Table IV. Mockup Rotary B₄C Control Rod Experiment

Position of $2 \times \frac{1}{2} \times 16$ -in. Column	Worth Depleted Uranium (4.7 kg) Relative to Void (lh)	Worth Enriched B ₄ C ^(a) [0.518 kg ^(b)] Relative to Void (lh)	Worth Enriched B ₄ C ^(a) Relative to Depleted Uranium (lh)
$\frac{1}{2}$ in. from core	+2.1	-36.9	-38.0
6 in. from core	+0.5	-1.9	-2.4

(a) Enriched B₄C = 69.3 w/o B; B = 90.7 a/o B¹⁰.

(b) Equivalent to 20.3 gm B¹⁰ per inch of axial length.

f. Axial Blanket Experiments - Assembly 41 resembles closely in composition an exponential experiment performed with ZPR-IV.² In the exponential experiment, measurements were made of the reflector savings given by depleted uranium, stainless steel, aluminum, and lead. The core-edge reactivity coefficients for these materials in Assembly 41 (see Table III) show approximately the same relative worths between materials as was obtained for the reflectors in the exponential experiment. However, results with full-scale blankets and reflectors in Assembly 41 give differences in relative worth for aluminum. (Only steel and aluminum are in sufficient supply at ZPR-III for blanket reflector experiments; no experiments with lead were performed.)

Experiments with axial blankets of various combinations of depleted uranium, steel, and aluminum were carried out behind the core in Half No. 1 of the assembly. A 41-v/o Al, 43-v/o depleted U, 9-v/o steel blanket composition was constructed in the entire 12-in.-length x 34-in.-diameter axial blanket by replacing the depleted uranium in about half the regular axial blanket drawers in Half No. 1 with 100% aluminum (obtaining, thereby, a depleted uranium/aluminum checkerboard pattern). The substitution resulted in a reactivity loss of -15.8 inhours. A full-density aluminum blanket (containing about 80% aluminum and 10% steel by volume) in 49 axial blanket drawers behind a wedge-shaped sector of the core was found to be worth -13.5 inhours relative to the depleted uranium type.

In the steel blanket experiments, a 50-v/o steel, 40-v/o depleted uranium blanket obtained by substitution of steel for depleted uranium in half the axial blanket drawers of Half No. 1 was 96.4 inhours more reactive than the regular depleted uranium blanket. Forty-nine drawers containing all steel (about 93 v/o) in a wedge sector was found to be 69.8 inhours more reactive when replacing depleted uranium blanket.

²Beyer, F. C. et al., The Fast Exponential Experiment, Proceedings of the United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland, 5, 342 (1955).

The above data are recompiled in Table V, in which the worths of complete full- and half-density aluminum and steel axial blankets behind one end of the core are presented.

Table V. Reactivity Worths of Axial Blankets, 12-in. Axial Length x 34-in. Diameter (177 Drawers)

Blanket Type	Composition, Vol. %			Worth Relative to Depleted Uranium Type (1h)
	Depleted Uranium	Steel	Aluminum	
Depleted Uranium	84	9	-	0
$\frac{1}{2}$ -density Al	43	9	41	-15.8
Full-density Al	-	9	84	-48.7
$\frac{1}{2}$ -density Steel	43	50	-	96.4
Full-density Steel	-	93	-	252

g. Radial Reflector Experiments - Experiments are proceeding using various thicknesses of full-density aluminum and steel radial reflectors to replace normal depleted uranium in the radial blanket. Results with aluminum indicate an initial positive worth with a subsequent change to negative worth as reflector thickness is increased.

2. Preparations for ZPR-VI and ZPR-IX

a. Building - The installation of the control wiring for the reactor cell air-conditioning system has been completed, and preoperational testing of the system is in progress.

The installation of the argon gas-storage system is being completed.

b. ZPR-VI Assembly - The dual-purpose rod drive mechanisms are being aligned with the matrix tubes for loaded conditions. After this work is completed, the core and blanket regions will be loaded with structural, coolant, and blanket materials for the first assembly to be investigated. The fuel will be loaded after approval of the hazards report is received.

Detailed design drawings have been completed for an automatic, foil sample-changer unit, and component drawings for an oscillator are in the shops for fabrication.

c. ZPR-IX - The installation of the control wiring is approximately 90% complete. Upon completion of this work, preoperational testing of the control console and interlock circuitry will commence.

The aluminum matrix tubes have been stress relieved by the vendor and are to be returned to the Laboratory. The Laboratory Shops will redraw the tubes to meet specifications.

3. AFSR Experiments

a. Reactivity Meter - The oscillator element was installed in the beam hole. Reactivity was readjusted so that overall available excess reactivity with the oscillator in its most reactive position was about 60 inhours. A static calibration of the oscillator was then made as a function of angular position and the signal shape was found to be satisfactory.

A preliminary trial of the reactivity meter was made. Noise obscured the signal except when reactivity was fairly close to delayed critical. To correct this, it was decided to construct a more effective filter rather than to obtain a more sensitive chamber, which would increase the noise signal. A synchronous filter, consisting of an array of capacitors on a rotating disc, is being built. The capacitors form a rudimentary analog memory capable of integrating a number of cycles and should result in much improved signal-to-noise ratios.

b. Color Discrimination between Neutrons and Gamma - A set of matched scintillators has been ordered. They arrived damaged and were returned to the vendor for replacement. A matched set of glass color filters has also been received.

c. Interlaboratory Comparison of Counting Techniques - On August 14, a number of sets of uranium foils were irradiated and sent to Argonne, Hanford, and Los Alamos for comparison of fission counting techniques. Most of the results have been received and are listed in Table VI for a preliminary comparison.

Table VI Preliminary Summary of Analysis of Foils Irradiated August 14 1962 in AFSR

Laboratory	Foil No	Hole Position	Data	Fissions/gm of foil
Argonne	1 (U235)	Glory	Fissions/gm	$1.17 \times 10^{12} \pm 3''$
	2 (U235)	Glory	Fissions/gm	$1.10 \times 10^{12} \pm 3''$
Hanford	14 (U235)	Glory	$\mu\text{C Ba140/gm}$	1.06×10^{12}
	15 (U235)	Glory	$\mu\text{C Ba140/gm}$	1.12×10^{12}
	14 (U235)	Glory	$\mu\text{C Mo99/gm}$	1.02×10^{12}
Los Alamos	5 (U235)	Glory	(nvt σ_f) U235 and U238	1.19×10^{12}
	5 (U235)	Glory	Fissions/gm	1.164×10^{12}
Argonne	61 (U235)	Glory	Fissions/gm	1.73×10^{11}
	62 (U235)	Glory	Fissions/gm	1.71×10^{11}
Hanford	68 (U235)	Glory	$\mu\text{C Ba140/gm}$	1.62×10^{11}
	68 (U235)	Glory	$\mu\text{C Ba140/gm}$ (chemical process altered)	1.67×10^{11}
	69 (U235)	Glory	$\mu\text{C Ba140/gm}$	1.69×10^{11}
Los Alamos	64 (U235)	Glory	(nvt σ_f)	1.75×10^{11}
	64 (U235)	Glory	Fissions/gm	1.743×10^{11}
Argonne	3 (U235)	Graphite #1	Fissions/gm	$1.02 \times 10^{12} \pm 3''$
	4 (U235)	Graphite #1	Fissions/gm	$1.01 \times 10^{12} \pm 3''$
Hanford	16 (U235)	Graphite #1	$\mu\text{C Ba140/gm}$	1.09×10^{12}
	16 (U235)	Graphite #1	$\mu\text{C Ba140/gm}$ (chemical process altered)	1.07×10^{12}
	17 (U235)	Graphite #1	$\mu\text{C Ba140/gm}$	1.12×10^{12}
Los Alamos	16 (U235)	Graphite #1	$\mu\text{C Mo99/gm}$	1.02×10^{12}
	9 (U235)	Graphite #1	Fissions/gm	1.052×10^{12}

B. EBR-I

Encapsulated single crystals of copper and silver, and tensile specimens of SA 212B steel were irradiated for exposures varying from 1 to 44.4 Mw-hr. These irradiations are part of a series designed to study the irradiation effects produced by a high-energy neutron flux.

Danger coefficients of specimens of tantalum and rhenium were measured for the General Electric Company. The measurement was made 1.7 in. from the reactor center and yielded the following information: tantalum sample, 2.07 gm (over a 6-in. length), -0.7 ± 0.4 inhour; rhenium sample, 23.7 gm (over a 6-in. length), -1.6 ± 0.2 inhours.

Calibration data for the fission break monitor were taken with the addition of a known weight and surface area of U^{235} foils contained in a vented capsule in the core and exposed to the reactor coolant.

With the reactor shut down, neutron counts were measured with the antimony source both inserted and removed from the beryllium source thimble. Measurements were made in the graphite reflector and in the core. The count ratio with the antimony source "in" and "withdrawn" agreed for both positions at an average value of 13:1.

Following completion of the above experiments, the reactor was shut down and unloading of the Mark III fuel elements was begun. At shut-down, the Mark III core had been operated for 3220 Mw-hr.

The reactor oscillating equipment has been set up and checked out in preparation for the Mark IV loading.

The plutonium fuel for Mark IV has been received at the reactor site, together with the hardware and foils for measuring breeding gain. Representative sample fuel elements will be checked for thermal integrity of the NaK bond with eddy current test equipment.

C. EBR-II

1. Reactor Plant

a. Rotating Plug - The modifications to the rotating plugs and the freeze seals have been completed, and both rotating plugs have been reinstalled without particular difficulty on the primary tank cover (see Progress Reports for August, 1962, ANL-6610, and for September, 1962, ANL-6619, p. 16). The fuel-transfer mechanisms are yet to be installed on the plugs.

Additional wireways to cover the dip-ring heater holes and to protect the heater leads were installed on both plugs. Dust covers, to protect areas where the rotating-plug ring gears are exposed, were added.

b. Transfer Arm Mechanism Modification - A project to modify the transfer arm mechanism was described in ANL-6619 (Progress Report for September, 1962, p. 17). Thus far, the vertical shaft and the horizontal arm with the locking mechanism inside; the counter balance and the pedestal platform; the vertical shaft seal, the spline shaft and ball nut, and the assembly for manual rotation; and the lock actuating assembly have been modified, preassembled, and tested on a temporary 20-ft-high support structure. After satisfactory operation at Argonne, Illinois, these parts were shipped to the EBR-II site in Idaho to be installed in the primary tank. The assembly of these parts on the 20-ft support structure are shown in Figure 1.

Figure 1. Transfer Arm Mechanism for Test
on 20-ft-high Support Stand

The counter weight assembly, which comprises a group of new parts, is being preassembled at Argonne, Illinois, and will be shipped to Idaho by the end of the month.

c. Fuel-unloading Machine - The fuel-unloading machine, illustrated in Figure 2, transfers fuel subassemblies between the primary tank and a mobile interbuilding coffin which is used to move fuel subassemblies into or out of the reactor plant. The fuel-unloading machine consists of an upright, shielded storage coffin mounted on a self-propelled carriage, which travels on tracks from the transfer port on the primary tank to the interbuilding coffin. A mechanism, located on top of the 15-ft-high shielded coffin, drives the gripping device 27 ft in the vertical direction by means of a special folding chain. Thus, the gripping device can be lowered into the primary tank or into the interbuilding coffin, grip a fuel subassembly by its upper adapter, and move it into or out of the fuel-unloading machine. The bottom of the shielded coffin is closed by a rotating gate. During vertical movement of the gripper the bottom of the shield coffin is connected to the transport port or the interbuilding coffin by a pneumatically operated seal ring. An argon-circulation system allows cooling or heating of fuel subassemblies at various phases of the transfer sequence. Electrical controls and interlocks tie the operation of the fuel-unloading machine to the reactor fuel-handling control console.

Figure 2. The EBR-II Shielded Fuel-unloading Machine for Transferring Fuel Subassemblies in the Reactor Building between the Reactor Primary Reactor Tank and the Interbuilding Coffin

The following subassemblies were preassembled at the DuPage site and checked or tested separately: the self-propelled carriage and platform structure, the shielded coffin, the gate and bottom seal ring, the moveable doughnut shield, the gripper drive, the control panel and electrical equipment cabinet, the sodium-vapor filter, "Molecular Sieves," and that portion of the cooling system which is part of the carriage.

After successfully performing limited tests of the assembled machine, it was disassembled into major components for shipment to Idaho.

Installation of the rails for the fuel unloading machine was completed. It is anticipated that assembly of the machine will start in the very near future. A temporary argon cooling system for initial operation of the fuel-unloading machine and transfer port is being installed.

2. Power Plant

During the month of October, as weather has permitted, work continued on the repair and strengthening of portions of the cooling tower.

Miscellaneous work inside the Power Plant was also performed. To allow operation with small heat loads, bypass lines around the No. 2 cooling water-circulating pump and from the No. 4 cooling tower riser to the cooling tower basin were installed.

3. Sodium Boiler Plant

Work continued on the following items: (1) insulation of piping and equipment; (2) painting and coating of piping and miscellaneous ferrous surfaces and structural steel; (3) installation of heater wire and thermocouples and their related wiring; and (4) cement finishing of walls.

Fabrication and installation of sheet metal flashing around piping and equipment was completed.

All craft work is now complete above the mezzanine and in the yard, and final cleanup of that area is now in progress. The Package IV contractor is currently scheduled to complete all work by November 1, 1962. At that time, ANL operating forces will start final checkout of the building.

4. Fuel-Cycle Facility

a. Construction and Installation - A diesel generator and associated switchgear have been installed on the service floor of the Fuel Cycle Facility to provide emergency power for the operation of essential equipment in this building. Operational testing of this equipment was started. At the completion of these tests, the equipment will be incorporated into the Fuel Cycle Facility power system.

To aid in the examination of cell equipment, monocular viewers are being provided for viewing through the shielding windows of the Air and Argon Cells. Monocular viewers of two focusing ranges are being made available.

With achievement of design flow rates and correct flow distribution, the testing and balancing of the ventilating system was completed. A minor leak in one of the shielding window tank units, discovered some time ago, was located and repaired.

Testing for across-the-seat leakage of the transfer lock valves was completed. Some of the valves are unsatisfactory and will require rework or replacement.

After modification, the lifting mechanism for the large transfer lock appears to be satisfactory. The lifting mechanism for the small transfer locks is also being modified.

The installation of master-slave manipulators in the air cell of the Fuel Cycle Facility has been completed. Two sealed master-slave manipulators have been received for argon cell installation, and consideration is being given to providing honed sleeves to allow later installation of additional manipulators of this type.

Procurement of fuel-element and assembly hardware for fuel-refabrication operations is nearing completion, and many of the components are ready for assembly. Some difficulty has been experienced in obtaining satisfactory castings of the element tip, assembly grids, and end fittings.

Final evaluation of data obtained from the argon cooling system shows the control valves to be 100 percent or more oversized. All control is derived from a valve-stem travel of 0.5 to 40 percent of the total travel, which is about 1 in.

A trial with these valves resulted in the first section of the cooling coil taking more than its share of the load. With only a small volume of Freon going through the lower coils, the back-pressure valves have operated at a very low rate, causing chatter. In two cases, pilot diaphragms have been broken.

Separation of mixed glass and fuel scrap from the injection-casting demolding operation is being investigated. Two approaches being considered are a milling operation, followed by vibratory screen separation, and a flotation method with mercury as the separating medium.

Preparations were made to radiograph the argon cell, air cell, and portions of the subcell areas. Three 5000-curie sources delivered from MTR the 29th of October will be used for this test. The sources will be handled by in-cell operating manipulators.

b. Development of Equipment - A 36-in.-diameter plug is being designed for addition to the removable 6-ft-diameter steel hatch in the Air Cell. This plug will be used to transfer large items which cannot be accommodated by the 6- and 24-in.-diameter plugs now available in this hatch but which do not require the full 6-ft-diameter hatch.

A scoop which will remove uranium precipitate from a round bottom crucible in the skull-reclamation furnace was received and tested at Argonne. Its operation was judged satisfactory for handling the uranium precipitate, which clumps into large aggregates.

5. Core II Fuel Development

a. Fuel Jacket Development - Two hundred feet of Nb-1 wt-% Zr alloy tubing, measuring 0.257 in. I.D. x 0.012 in. wall, have been received from the Superior Tube Co. The tubing was eddy current inspected, pressure tested at 4500 psi, cleaned, and leak checked with a helium mass spectrometer. All of the satisfactorily inspected tubing was suitable for jacketing irradiation test specimens.

Fifty feet (random lengths) of Nb-4 wt-% V alloy tubing, measuring 0.156 in. I.D. x 0.009 in. wall, has been received from the Westinghouse Electric Corp. The tubing was eddy current inspected, pressure tested at 7300 psi, cleaned, and leak checked with a helium mass spectrometer. All of the satisfactorily inspected tubing will be made available for jacketing irradiation test specimens.

Nb-4 wt-% V alloy bar stock (0.450 in. in diameter) is being fabricated to produce 0.176-in.-diameter rod for end plug stock for the above tubing.

b. Irradiation of Prototype EBR-II Core-II Fuel Elements - Post-irradiation examination of a group of specimens irradiated in CP-5 is presently being conducted. Two irradiation capsules, CP-22 and CP-23, have been opened and dimensional measurements of the specimens have been made. Each capsule contained six specimens of U-20 wt-% Pu-10 wt-% F's alloy clad with refractory metal alloy arranged in two tiers of three each. The discontinuation of the irradiation period was prompted by erratic temperature readings which suggested failure of some of the claddings. This was confirmed by neutron radiography of the capsules, which clearly identified the specimens which had failed.

The specimens in capsule CP-22 deviated from the reference EBR-II fuel-element design in that the sodium bond had been purposely omitted and the fuel was permitted to melt within the claddings during reactor startup. The specimens were irradiated to approximately 1 a/o burn-up, at which time there was evidence of cladding failure.

Visual examination of the specimens in the hot cell revealed that the three claddings in the top tier (Nb-1 wt-% Zr, Ta-0.1 wt-% W, and Nb-33 wt-% Ta-1 wt-% Zr) which were being irradiated at 690°C center fuel temperature had failed. The failures were characterized by longitudinal splits or ruptures at the midportion of the specimen out of which fuel material had extruded. The cladding which suffered the greatest damage was the Nb-1 wt-% Zr, which had a longitudinal split extending for one inch. The bottom-tier specimens in the capsule were being irradiated at 640°C center fuel temperature, and the cladding compositions were Nb-1 wt-% Zr, Nb-33 wt-% Ta-1 wt-% Zr, and vanadium. The Nb-1 wt-% Zr jacket had a longitudinal split which extended for 75% of the fueled portion of the specimen. The other two specimens were intact; however, the vanadium-clad specimen was bowed.

Capsule CP-23 contained specimens which were sodium bonded, but the cladding length had been increased 0.5 in. over the prototype clad length. This modification was made to reduce the gas pressure in the void space above the fuel so that at the start of irradiation these pressures were calculated not to exceed 50 psi. The capsule was removed from the reactor after a calculated 1.3 a/o burnup had been achieved and after erratic temperature readings indicated that specimen failures may have occurred. The claddings of the bottom tier (Nb-1 wt-% Zr, Ta-0.1 wt-% W, and Nb-33 wt-% Ta-1 wt-% Zr) which were irradiated at 660°C central fuel temperature were intact. Identical claddings were being irradiated on the top tier at 620°C central fuel temperatures. Of these, the Ta-0.1 wt-% W was intact. The other two had 0.5-in. splits in the claddings at the midplane of the fueled portion of the specimen.

Diameter measurements taken on all specimens at positions other than where the cladding ruptured failed to indicate any significant dimensional increases.

Transverse sections of U-20 wt-% Pu-10 wt-% Fs fuel pins which failed during irradiation were examined metallographically in an effort to determine the mechanisms by which the failures had occurred. The fuel specimens were canned in different refractory alloys (Inconel-X with $\frac{1}{2}$ mil vanadium barrier, niobium, vanadium, and niobium-1 wt-% zirconium alloy) in order to investigate the relative effectiveness of the various materials on restraining swelling of the fuel alloy. The assembled fuel pins had been irradiated in instrumented capsules in CP-5 to burnups of from 1.1 to 1.6 a/o and at central fuel and surface temperatures ranging from 730 to 1000°C and from 605 to 760°C, respectively.

There was no discernible diffusion zone between the core and the vanadium cladding in areas away from the failed area, and the $\frac{1}{2}$ -mil vanadium barrier prevented any interaction between the fuel and the Inconel-X cladding. Some interdiffusion was detected between the fuel and the niobium and niobium-1 wt-% zirconium alloys, although not to any great extent.

Cracks were found at the inner surfaces of the niobium-1 wt-% zirconium alloy claddings, but none could be found in either the vanadium, Inconel-X, or niobium-clad samples. The majority of the niobium-1 wt-% zirconium claddings appeared to have failed ultimately by shear. The cracks which developed as a result of the high hydrostatic pressures caused by fuel expansion apparently weakened the cladding to such an extent that the shear mechanism was able to come into effect. In a vanadium-clad sample in an area near where failure had occurred, a larger diffusion zone was found, apparently caused by the higher temperatures encountered in this region. Cracks which developed in this diffusion zone were at a 45° angle to the surface but did not penetrate into the cladding.

An examination of the areas of the niobium and niobium-1 wt-% zirconium claddings where failure had actually occurred showed that complete reaction had taken place between the core and the clad so that fissioning was observed in what once had been cladding. This had occurred to such an extent that the cladding was no longer distinguishable from the fuel, except in some small areas.

There was no particular pattern of void distribution within the fuel alloy of the samples. It was noted, however, that the voids in the fuel clad in niobium-1 wt-% zirconium alloy in an area where complete failure had occurred assumed a circular pattern running parallel to the outer perimeter of the specimen. It is possible that this particular distribution was influenced in some degree by the thermocouple hole which normally extended into this same area.

6. Process Development

a. Melt Refining Process Technology - Analytical results on the behavior of individual fission products have been received for the fifth small-scale, high-activity-level melt-refining experiment (see Reactor Development Program Progress Report, June 1962, ANL-6580, page 22). A 382-gm charge of 10 percent enriched uranium-5 percent fission alloy, irradiated to 1.75 total atom percent burnup and cooled 25 days, was melt refined under argon for one hour at 1400°C in a lime-stabilized zirconia crucible. This experiment differed from the four previous ones in two respects: (1) irradiation to a higher burnup (1.75 percent vs 0.22 to 0.87 percent), and (2) use of air oxidation, instead of organic solvents, to destroy residual NaK coolant on the fuel pins when they were removed from the irradiation capsule. There was no significant difference in the behavior of fission products between this experiment and the fourth high-activity-level melt-refining experiment. In these two experiments, uranium showed no preferential distribution to either the ingot or the skull, as compared with the distribution of the total metal in the charges;

molybdenum, a typical noble metal, showed a slight enrichment in the ingot; rare earths and yttrium distributed principally to the skull, with about 15 percent appearing in the crucible; tellurium appeared mainly in the skull, but about 25 percent of the tellurium in the charge was not accounted for; barium and strontium distributed primarily to the crucible; the zirconium showed some enrichment in the skull. The following variables had no significant effect on the fission product removals in the five runs: (1) irradiation level, (2) activity level, (3) fuel preparation procedure, and (4) duration of melt refining (one hour and three hours).

As part of an investigation to evaluate the use of fluxes in the melt refining process, several runs were completed in which 700-gm charges of uranium-5 percent fissium pins containing cerium (about 0.8 percent) were melt refined under argon in beryllia crucibles. Either uranium tetrachloride or magnesium fluoride was used as an oxidizing agent, and the flux generally used was calcium chloride. Liquation temperatures were 1100° to 1300°C, and the liquation time was one hour. For ingots poured at 1300°C, yields varied between 96 and 98 percent, and cerium decontamination averaged about 94 percent.

b. Skull-reclamation Process - Work is proceeding on the evaluation of pressed Alundum crucibles for use in the skull-reclamation process. A 300-gm uranium run was performed with a crucible which had a porosity of approximately 19 percent. Severe leakage of zinc-magnesium occurred during the intermetallic precipitation step and the uranium precipitation step. The portion of the material that penetrated the crucible wall was contained in a tantalum secondary crucible. In subsequent retorting, residual zinc and magnesium were removed and 246 gm of uranium was recovered. Now being considered for this application are Alundum crucibles formed by methods other than pressing and magnesia crucibles.

The skull-reclamation process requires the heating and mixing of molten metal-salt systems. Low-frequency inductive heating and mixing is being investigated for this purpose. The adequacy of 60-cycle power for melting $\frac{1}{2}$ -in.-diameter zinc rods was tested to determine whether rods may be used as a charge material. The temperature rise (to 335°C) obtained with a 78.5-lb charge of zinc rods indicated that successful melting is likely to occur with the larger power sources planned for the Fuel Cycle Facility. This would eliminate a zinc-premelt step in the skull-reclamation process. Inductive mixing experiments with magnesium or zinc have shown that, for the same power input, the agitation was 3 to 4 times more vigorous for magnesium than for zinc. This suggests that, as the proportion of magnesium increases in zinc-magnesium mixtures, an increase in the vigor of agitation should result.

c. Blanket-processing Studies - Four additional runs on a 4-kg uranium scale have been performed, completing the plant-scale demonstration of the phase-separation step which follows uranium precipitation in the

blanket process (see Reactor Development Program Progress Report, September 1962, ANL-6619, page 24). The results of nine runs show that it is possible to achieve transfer of 95 percent of the plutonium-bearing supernatant with less than one percent entrainment of uranium. This is regarded as satisfactory.

d. Plutonium Recovery Process - The distribution coefficients of praseodymium and plutonium between zinc-magnesium alloy and 50 mole percent magnesium chloride-barium chloride at 800°C were determined. Both distribution coefficients were lower when barium chloride was in the flux than for other flux compositions (see Reactor Development Program Progress Reports for August 1962, ANL-6610, page 35, and for September 1962, ANL-6619, page 24). It also was found that the plutonium-praseodymium separation factors ranged between 17 and 43, depending upon the magnesium concentration in the metal phase. Two runs were performed to determine the distribution behavior of iron (a possible constituent of cladding alloys) between zinc-magnesium alloys and magnesium chloride at 800°C under argon. Iron showed a preference for the zinc-magnesium phase. One run was conducted at 800°C under argon to determine the distribution behavior of vanadium (another possible constituent of cladding alloys) between zinc-5 wt-% magnesium alloy and magnesium chloride. Vanadium showed a preference for the alloy and was found to precipitate in the metal phase, apparently as a zinc-vanadium inter-metallic compound.

A process which has been suggested as an alternative to present methods of processing uranium-plutonium-fission alloy fuels for fast reactors involves oxidation of the uranium, plutonium, and rare earth fission products into a molten halide flux. The uranium and plutonium are then simultaneously reduced and coprecipitated by means of a calcium-rich zinc alloy. The rare earths are reduced, but remain in solution. In a preliminary demonstration of the procedure, 95 percent of the plutonium and virtually all of the uranium were precipitated. It was discovered, however, that some lithium and potassium from the lithium chloride-potassium chloride salt were also reduced, indicating that a more stable salt phase, such as calcium chloride, is necessary.

e. Materials and Equipment - Two high-density (about 93 percent of theoretical), pressed beryllia crucibles successfully contained calcium chloride-magnesium chloride-magnesium fluoride ($47\frac{1}{2}$, $47\frac{1}{2}$, and 5 mole percent, respectively) flux for 24 hr at 800°C without leakage of flux or crucible damage. Under the same conditions, two dense, high-fired alumina crucibles were permeated by the flux and developed cracks, perhaps due to thermal shock.

7. Training Program

The resumption of systems training, a new offering of a reactor technology course, and fire-protection training constituted training activities during this period.

Nineteen technicians completed a plant-orientation phase of systems training. This effort involved some 800 man-hours of guided classroom study of the Hazards Report and Addendum, selected operating manuals, and field sessions in the Power Plant, Sodium Boiler Building, and Reactor Plant.

Systems training is proceeding according to a formal operator-qualification outline drawn up by the staff of the operations group.

This program identifies facility orientation, plant orientation, and radiation protection and plant safety as training areas for all operators by means of group lectures, individual study, and field sessions. Other phases to complete the preparation for qualification are training in a specific assigned system, overall plant operation, and actual experience on an operable system.

Ten EBR-II technicians, together with four assigned to the BORAX-V, EBR-I, and AFSR reactors, participated in eight daily 3-hr sessions of a reactor technology course.

Twelve technicians spent five one-hour daily sessions in fire-protection safety training exercises conducted by a member of the Argonne, Illinois, Fire Department.

D. FARET

An experimental program to be performed in the FARET facility is being developed. The program is divided into the following two major areas: (1) high-performance engineering tests, and (2) physics and dynamic tests.

It has been proposed that a small volume core should be used for the high-performance engineering tests, in order to achieve a high power density in the core within the limitation of the total 50-Mwt power capability of the FARET facility. Two cores with volumes of 50 liters and 25 liters, respectively, were investigated. The fuel, coolant, and structure used were PuC-UC, sodium, and niobium in the volume ratio of 30/52/18, respectively. The physics and engineering parameters calculated are listed below:

Core Size (liters)	L/D	Plutonium Enrichment	Critical Mass of Plutonium (kg)	Power Density (Mw/liter)	Specific Power of Plutonium, (Mw/kg)	Heat Rating, PuC-UC (Mw/kg)
50	1.0	0.42	85.8	1.0	0.583	0.245
25	1.0	0.65	66.3	2.0	0.754	0.490

The capability for heat removal is within the limitations of the existing sodium technology. For the 25-liter core the value of $v\Delta t$ for the coolant is approximately 9000 ft-°F/sec, which corresponds to a flow velocity of 30 ft/sec and a temperature rise in the core of 300°F.

For these small cores the sodium-temperature coefficient and the fuel-expansion coefficient were also calculated. The values are:

Core Size (liters)	Sodium Void Coefficient (dk/dt/°C)	Fuel Expansion Coefficient (dk/dt/°C)
50	-5.4×10^{-6}	-4.9×10^{-6}
25	-7.3×10^{-6}	-5.1×10^{-6}

There is some question about the reliability of the fuel expansion of ceramic fuels available in these cores. A small core with part ceramic and part metal fuel loading is now being considered. The metal fuel with its reliable fuel expansion will provide the necessary prompt negative reactivity feedback for the safe operation of this core. The Doppler temperature coefficient in these small cores has been estimated to be negative, but the value is, of course, very small.

A medium-sized core has been proposed for use in the physics and dynamic tests with the FARET facility system. In this system, the various temperature coefficients of reactivity will be determined by nuclear heating of the full core. A core size of 400 liters and a composition of PuC-UC/Na/Nb, 30/52/18 by volume percent was investigated for this purpose. The estimated magnitude of the Doppler effect on reactivity for a change in temperature from 750°K to 1500°K is:

Core Size (liters)	Ratio Pu/U ²³⁸	Doppler Δk 1500°K 750°K	Doppler (dk/dt/°C)
400	1:4	-0.001155	-1.54×10^{-6}

The sodium and fuel-expansion temperature coefficients are dependent on the L/D ratio for this core. Calculations were performed for the above core with L/D ratios of 1.0, 0.5, and 0.25. The results are:

Core Size (liters)	L/D	Sodium Void (dk/dt/°C)	Fuel Expansion (dk/dt/°C)
400	1.0	-1.0×10^{-6}	-4.83×10^{-6}
400	0.5	-1.4×10^{-6}	-3.16×10^{-6}
400	0.25	-2.75×10^{-6}	-1.34×10^{-6}

The fuel-expansion effect on the reactivity is smallest for a small ratio of L/D. Thus, the experiment on the determination of Doppler effect could best be done with a core with L/D = 0.25 in order to reduce the effect of the competitive fuel expansion.

III. GENERAL REACTOR TECHNOLOGY

A. Applied Reactor Physics

1. Measurements of Fast-neutron Spectrum

Part of the current program of measurement of fast-neutron spectra is directed towards improving the interpretation of pulse-profile structure from a proportional counter filled with hydrogen gas and to develop proton-recoil detectors suitable for use in fast reactor spectroscopy in the energy region below 200 kev.

Some results from the proportional-counter-recoil-proton neutron-spectroscopy program in which a fission converter plate was used were reported in ANL-6544 (Reactor Development Progress Report for March, 1962, p. 32). Pulse-profile discrimination was used to effect a substantial reduction in gamma-ray sensitivity and permitted the recoil-proton spectra to be measured at energies lower than would otherwise be possible.

More recent studies have been made that indicate that the low-energy response of proton-recoil proportional counters can be extended by a decade or so, except in those instances where gamma background is too intense to be removed by pulse-profile discrimination. It is also of considerable interest to determine under what conditions, if any, it is possible to introduce a proportional-counter probe of minimal size into a fast assembly in order to determine the spectroscopy in the energy region below 200 kev (that part of the spectrum above 200 kev will probably be successfully studied by other techniques, including the newly developed solid-state lithium sandwiches). Indications are that hydrogen gas at a pressure of a few atmospheres would be used and that wall effects due to proton recoils from the more energetic end of the spectrum will cause a serious distortion in the region below 200 kev, where studies are to be emphasized.

There is a possibility, however, that these wall effects can be identified through the analysis of the profile and rejected. Wall effects are expected to produce pulses which rise more slowly than pulses arising from normal recoils for the same total ionization. This difference may be sufficiently great to permit discrimination between the desired and undesired pulses.

2. High-conversion Critical Experiment: ZPR-VII

The study of the properties of moderately enriched uranium, light-water-moderated reactors has been resumed in the ZPR-VII facility. Assemblies having a fuel-to-water ratio higher than heretofore studied are of particular interest because of the high conversion ratio as indicated by theoretical studies of such systems and the high fuel-to-water volume ratios which are characteristic of typical designs of boiling water reactors.

A detailed investigation of uniform lattice assemblies using 5 wt-% enriched uranium, stainless steel-clad BORAX-V boiling zone fuel pins with hydrogen-to- U^{238} atom ratios of 3.5 and 4.7 have been made. The current assembly consists of the 3 wt-% enriched uranium, stainless steel-clad Hi-C fuel pins in a 1.24-cm square pitch pattern. The fuel is in the form of 0.935-cm-diameter pellets of 10.2-gm/cm^3 density, stacked to a height of 122 cm inside a 1.06-cm-diameter, Type 304 stainless steel tube with a 0.10-cm wall thickness. At the 1.24-cm spacing the core has a hydrogen-to- U^{238} atom ratio of 3.0, equivalent to a water-to-fuel volume ratio of unity.

The customary loading to criticality, control rod calibration, and reactivity measurements have indicated that 1765 fuel pins are required for a fully reflected, cylindrical critical assembly containing no control rods or channels within the core. It is of interest to note that the corresponding number of aluminum-clad Hi-C fuel pins is approximately 990, a value derived from some measurements made at the start of the Hi-C program. The top reflector of 20-cm-thick water has a total worth of $0.115\% \Delta k/k$, equal to the reactivity effect of 16 peripheral fuel pins.

A 60-cm-diameter cylindrical loading of 1750 fuel pins has been established as the base for determining the various reactor parameters, such as reflector savings, resonance escape probability, fast effect, thermal utilization, and neutron spectrum. The reactivity of the assembly is controlled by movement of the three five-element- B_4C -loaded finger rods installed in the core and four boral blade rods installed in the radial reflector. The finger rods are each worth $0.71\% \Delta k/k$, the blade rods $0.24\% \Delta k/k$ each in reactivity.

3. Absolute Measurement of ν for U^{235} and Cf^{252}

An improved measurement for ν , the average number of neutrons per fission, of U^{235} and Cf^{252} is currently being pursued by groups at Aldermanston, England, the Nobel Institute in Sweden, and Los Alamos, as well as at ANL. The other groups are performing coincidence-type experiments with large liquid scintillators for neutron detection. The accuracy of this method depends upon a difficult calibration of the response of the liquid scintillators to a fission spectrum and stability of an elaborate counting system throughout the measurement. The scintillators must be calibrated over a spectrum of discrete energy levels by means of accelerator-produced neutrons.

A simpler method based upon absolute measurement is being developed here. The emitted neutrons from the fissile sample incorporated in a fission counter are counted indirectly by allowing them to activate manganese dissolved as a compound in water. The disintegration rate in a liquid sample can be determined with relative ease and the measurement can be performed frequently. The accuracy of the measurement can be

made to be only slightly dependent on the efficiency of the various counting channels by using coincidence techniques. The absolute count of fissions can be made by a separate calibration of the fission counts in coincidence stability. The value of ν can be determined from these measurements.

During 1961, the various components required for this experiment were built and tested; the first experiments were performed with the CP-5 as a source of thermal neutrons to irradiate the U^{235} fission counter. Analysis of the data showed, however, that the liquid sample counter was not sufficiently stable (see ANL-6485, Reactor Development Program Progress Report, December, 1961, p. 41). The counter was redesigned and new electronics devised. The hardware for this new equipment and some of the circuitry was completed last spring (see ANL-6565, Reactor Development Program Progress Report, April, 1962, p. 34). Meanwhile, different schemes for counting the disintegrations, having 100% efficiency, were devised and were tested with the coincidence calibration equipment. As a result of these tests, a two-tube coincidence counter was built that had a satisfactory degree of stability.

Another phase of this experiment concerns the development of a fast and efficient channel to detect fission neutrons used in calibrating the fission counter. This channel must be highly effective in the discrimination against gamma radiation. Efforts to improve the gamma-ray discrimination have led to the development of a special pulse-shape discriminating circuit for Hornyak-button neutron detectors, and an investigation of the decay of zinc-sulfur scintillations, as well as the scintillations from a number of other materials.

The solution counter and coincidence equipment are expected to be ready next month; the fission channel calibration equipment should be in operating condition in the following month.

4. Elastic Scattering of Neutrons

a. Scattering from Iron - The analysis of elastic-scattering measurements with iron is completed. The angular distributions have been fitted to a Legendre polynomial expansion of the form

$$\frac{d\sigma_{el}}{d\Omega} = \frac{\sigma_{el}}{4\pi} \sum_0^4 \omega_i P_i(\cos \theta); \omega_0 = 1 \quad (1)$$

The data are fully corrected for effects due to multiple scattering, attenuation of neutron flux in the scattering sample, and finite angular resolution of the detector. The results given in Figure 3 include the Legendre polynomial coefficients $\omega_1, \omega_2, \omega_3$, and ω_4 ; the integral elastic-neutron-scattering cross

section σ_{el} ; and the total neutron cross section σ_t . The iron data were obtained by means of the neutron gamma-ray discrimination detector with an incident-neutron-energy resolution of approximately 20 kev.

Figure 3. Elastic Scattering of Neutrons in Natural Iron

b. Elastic Scattering from Natural Tungsten - The measurement of the differential scattering cross section of natural tungsten has been completed from an incident neutron energy of 300 kev to 1.5 Mev. Fast time-of-flight techniques provided scattered-neutron resolutions of 20 kev at 500 kev and of 150 kev at 1.5 Mev. The experimental results were corrected for all experimental perturbations, including the effect of multiple scattering within the samples. The measured differential cross section is given in Figure 4 in the form

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_{el}}{4\pi} \left(1 + \sum_{i=1}^5 W_i P_i \right) , \quad (2)$$

where P_i are Legendre polynomials, W_i experimentally determined constants, and σ_{el} the total elastic cross section. The solid data points refer to experimental measurements carried out with sufficient spectrometric resolution to resolve clearly the elastically scattered neutron group from the accompanying inelastic components. The open circular points refer to measurements which included the first inelastic component ($Q = -115$ kev) with the elastically scattered neutrons. These latter measurements were corrected for this inelastic contribution and the results plotted as the open triangles shown on the figure. The corrections were obtained from measured inelastic cross sections at selected angles between 1.3 and 1.5 Mev.

Figure 4. Elastic Scattering of Neutrons in Natural Tungsten

c. The Elastic Scattering of Fast Neutrons from U^{238} - Concurrently with the measurement of the elastic-scattering cross section of natural tungsten described above, the determination of a differential elastic-scattering cross section of U^{238} was carried out. The technique employed was identical with that outlined in the previous section, and the final results are obtained in the form of Eq. (2) above. The differential elastic-scattering cross section of U^{238} so represented is shown in Figure 5. As was the case with natural tungsten, the experimental resolution was not sufficient to distinguish clearly between the elastically and inelastically

scattered neutrons throughout the incident neutron energy range from 0.3 to 1.5 Mev. In the figure, the solid data points denote measurements that clearly resolved elastically scattered from inelastically scattered neutrons. The open circles represent measurements that included the first inelastic component ($Q = -45$ kev) with the elastic group. The square points denote measurements including both the first and second inelastic group ($Q = -45, Q = -150$ kev) with the elastic component. In all instances, the measurements containing inelastic contributions were corrected to the elastic component. These corrected values are indicated by the open triangles.

Figure 5. Elastic Scattering of Neutrons in U^{238}

5. Spontaneous Fission Spectrum of Cf^{248}

The kinetic-energy distribution of fragments from spontaneous fission of Cf^{248} was measured relative to Cf^{252} . The preliminary results are:

	<u>Cf²⁴⁸</u> <u>(Mev)</u>	<u>Cf²⁵²</u> <u>(Mev)</u>
Most probable energy of light fragment	110 ± 1	107
Most probable energy of heavy fragment	81 ± 1	79
Most probable total energy	191 ± 1.4	186

6. Theoretical Physics

a. Numerical Methods Analysis - The efficiency of various methods of estimating integrals of functions of several independent variables was compared by experimental calculations with the LGP-30 computer. Three methods of quadrature were investigated: (1) simple Monte Carlo, in which the estimate is the mean of N values of the integrand evaluated at randomly selected points in the region, multiplied by the volume of the region; (2) the composite formula (see Reactor Development Program Progress Report, Sept 1962, p 31); (3) the Cartesian product of one-dimensional Gaussian formulas (i.e., use of a Gaussian formula to estimate the integral over each variable independently). These formulas are typical of integrating over the entire region with a formula which is exact for polynomials of relatively high degree, although they are more effective than many such formulas for integrands which depend only upon the product of the independent variables.

All integrals were estimated over the four-dimensional hypercube $0 \leq x^{(i)} \leq 1$ ($i=1,2,3,4$). The following integrands were used:

$$(1) \quad \int_V \exp \left(x^{(1)} x^{(2)} x^{(3)} x^{(4)} \right) dV = 1.0693976 \quad ;$$

$$(2) \quad \int_V \sum_{i=1}^4 \left| x^{(i)} - \frac{1}{2} \right| dV = 1.0000000 \quad ;$$

and

$$(3) \quad k \int_V (\cos w - 7 w \sin w - 6 w^2 \cos w + w^3 \sin w) dV = \sin k \quad ,$$

where

$$w = k x^{(1)} x^{(2)} x^{(3)} x^{(4)} \quad .$$

Typical results are shown in Figure 6, in which the magnitude of the error is plotted against the number of function values used. The errors for the Monte Carlo estimates are the standard deviations of sets of five independent estimates. There is approximately one chance in three that this error will be exceeded by a single estimate. It will be observed that, except for Method (2), for which any composite formula with the ranges divided into an even number of subdivisions happens to be exact, but which is also the only integrand with a singularity, the Gaussian formulas are the most efficient. Monte Carlo is the most efficient only if the desired precision is relatively low.

Figure 6. Relationship of Error Magnitude with the Function of the Values Used in the Calculation

b. Theoretical Studies for ZPR-III - Examinations of the potentialities of the two-zone loading, discussed in ANL-6619 (Reactor Development Program Progress Report for September, 1962, p. 30), with the central zone characteristic of a large power breeder has continued. Cylindrical calculations indicate that the two-zone radial real and adjoint fluxes are flatter than for the full-sized core. The most important consequence of this result is that Doppler studies which might be disturbed by reactivity changes due to radial thermal expansions in the full-sized core can be more easily performed in the two-zone arrangement.

A particular study of interest is a system which may be experimentally investigated as both a single-zone and a two-zone system as a means of verifying the accuracy of the two-zone method. Since the size limitation is established by materials currently available at the ZPR-III facility, a two-zone system having the composition of Assembly 34, a dilute carbide system which was previously investigated, may be selected for this study.

B. Reactor Fuels and Materials Development

1. Corrosion Studies

a. Niobium Alloys - Alloys of vacuum annealed, as-received, Nb-1.84 wt-% Cr and Nb-4.33 wt-% Zr showed a constant microhardness across their transverse sections in contrast with the large increase in surface hardness of the respective alloys tested in flowing sodium (reported to contain 1-5 ppm Na₂O). The hardness at the center of the specimens exposed to sodium corresponds to the hardness of the vacuum-annealed specimen, indicating that no hardness change resulted solely from the time spent at temperature.

As illustrated in Figures 7 and 8, the increase in hardness of the samples tested in sodium extended over an appreciable fraction of the cross section. It is possible that this increase in hardness could be accompanied by a loss of ductility. This possibility will be investigated.

b. Lightweight Alloy for Use with Mercury - It was previously found that the solubility of aluminum in mercury is high and is temperature dependent. Large amounts of aluminum in titanium often promote an accelerated corrosion attack in high-temperature mercury.

Nitrogen and aluminum are alpha formers in titanium alloys. It has been shown that the formation of titanium nitride on a titanium surface protects the material from mercury attack. During this reporting period, interest has been directed to the possibility of replacing aluminum with nitrogen and to investigate its effect on mercury corrosion of some titanium alloys. Nitrogen is added as titanium nitride (TiN).

Figure 7. Hardness Profile of Nb-4.33 wt-% Zr Specimen in Flowing Sodium

Figure 8. Hardness Profile of Nb-1.84 wt-% Cr Specimen in Flowing Sodium

A series of alloys has been prepared, and corrosion tests have been performed for a period of 336 hr in mercury at 538°C. Test results are shown below:

Corrosion of Titanium Alloys in Mercury at 538°C Static

Aluminum-containing titanium alloys	Weight change (mg/cm ²)	Nitrogen-containing titanium alloys	Weight change (mg/cm ²)
Ti-2.5 wt-% Al-16 wt-% V	-21.47	Ti-0.12 wt-% N-16 wt-% V	- 8.65
Ti-3 wt-% Al-5 wt-% Cr	-53.90	Ti-0.15 wt-% N-5 wt-% Cr	-22.90
Ti-7 wt-% Al-12 wt-% Zr	-77.56	Ti-0.35 wt-% N-12 wt-% Zr	-25.05

The new ternary alloys containing nitrogen were improved in corrosion resistance by a factor of approximately three over the aluminum-containing alloys of comparable composition. Metallographic examinations of these alloys are now in progress.

2. Ceramic Fuels

a. Urania-Thoria Solid Solutions - As reported previously (Reactor Development Program Progress Report, November 1961, ANL 6473, p. 40), an anomalous behavior in the thermal expansion curves of nonstoichiometric solid solutions of urania-thoria heated in vacuum suggested an oxygen loss in the solid solutions. Stoichiometric and nonstoichiometric ratios of urania and thoria were prepared for differential thermal analysis to study the oxidation behavior of the materials.

The raw materials for the study were U_3O_8 and ThO_2 , calcined at 1200°C in air for 8 hr. Fused ThO_2 was the standard in the differential thermal analysis (DTA) for all determinations.

The thermogram for freshly calcined U_3O_8 showed small endothermal peaks at 145° to 160°C , at 230°C , and at 310°C , and a major endotherm at 920°C . The latter peak is the reduction of U_3O_8 to a lower oxide form, probably UO_2 . The nature of the reaction causing the lower temperature endotherms has not been determined. These endotherms were evident in mixtures ranging from 2.5 m/o UO_2 to 90 m/o UO_2 . Solid-solution formation began around 1000°C and reached completion around 1200°C , as indicated by a broad endotherm in this temperature region.

Other thermograms were made on mixtures which had been prepared approximately six months prior to these determinations. In addition to the endotherms found in the freshly calcined materials, an exotherm around 350°C appeared only in the mixtures of urania and thoria. The DTA curve for the aged urania showed no exotherms.

Endotherms may be associated with phase changes, with oxygen or water losses, or with solid-solution formation. Exotherms may be associated with oxidation or phase changes.

b. TTA Complexes - Single crystals of the TTA chelates of tetravalent uranium, thorium, hafnium, zirconium, cerium, and plutonium have been prepared and examined by single-crystal X-ray techniques. X-ray diffraction data revealed that the compounds are isostructural with only slight variations in the orthorhombic cell dimensions. The variations are thought to stem from the differences in the covalent radii of the metals. Studies were made of the optical properties, infrared absorption spectra, and DTA curves of the compounds. Melting point data were obtained from the DTA curves.

Studies are currently being conducted with crystalline $\text{UO}_2(\text{TTA})_2$. Preliminary results show that there are two modifications of this compound, one having a cell size exactly twice that of the other. The form with the larger unit cell is obtained by crystallizing from benzene. It is red in color, exhibits marked pleochroism, and has the orthorhombic cell dimensions: $a = 11.4 \text{ \AA}$, $b = 51.2 \text{ \AA}$, and $c = 7.4 \text{ \AA}$. The other form is derived by crystallizing from ethyl alcohol. It is yellow to orange, also exhibits pronounced pleochroism, and has these unit cell dimensions: $a = 11.4 \text{ \AA}$, $b = 25.6 \text{ \AA}$, and $c = 7.4 \text{ \AA}$.

The yellow crystals of uranium peroxy-oxalate $[(\text{NH}_4)_2 \text{U}_2\text{O}_8 \text{C}_2\text{O}_4 \cdot 4\text{H}_2\text{O}]$ are monoclinic, with the following unit cell dimensions: $a = 27.8 \text{ \AA}$, $b = 20.1 \text{ \AA}$, $c = 10.6 \text{ \AA}$, and $\beta = 99^\circ 36'$. These crystals suffer crystalline degradation when exposed to X-rays. After exposure of 30 to 50 hr to unfiltered Cu radiation, the crystals were virtually amorphous and gave a

diffuse diffraction pattern. This behavior is thought to be associated with destruction of the peroxide bonds.

c. Uranium Phosphide - The objective of this study, as described in the July report, is to characterize the various compounds in the uranium-phosphorus binary system and evaluate their potential as reactor fuel materials. Compounds are being made by solid-solid reaction of the elements, although other methods of synthesis will be investigated.

Chemical analysis of uranium phosphide material, which exhibited a melting point of 2640°C , showed the following major constituents: $\text{U} = 88.75\%$, $\text{P} = 10.0\%$, and $\text{O} = 1.25\%$. This analysis corresponds to a UP content of 88.2% , the remaining 11.8% being UO_2 . Similar material was arc melted and showed no change in melting point. Presumably this indicated an unchanged composition as a result of arc melting. UP and UO_2 appear to be chemically compatible with each other, even above the melting point of UP. Efforts are being made to reduce the oxygen content of the material in order to obtain a pure UP sample for more accurate evaluation of its properties.

Melting point determinations of U_3P_4 , which contained about 11% of UO_2 , were made. The material was considerably darker than UP. It appeared to undergo some changes at 2400°C and finally melted at about 2600°C . X-ray analysis of this material, heated just below 2600°C , showed it to consist of UP and small amounts of UO_2 . The material appeared compatible with the tungsten boat. Apparently, U_3P_4 dissociates in the vicinity of 2400°C to give UP and P, the latter volatilizing readily. These results are preliminary and phase studies of the system U-P will begin once compounds of higher purity are made or received.

3. Nondestructive Testing

a. Ultrasonic Techniques

i. Correlation of heat transfer properties and bond quality - This program has been formulated for the purpose of providing a partially quantitative definition of the relationship between bond quality and heat transfer properties. The assumption in work of this kind has always been that poor bonding of a sufficiently large area of cladding, as indicated by the ultrasonic tests, provided sufficient grounds for rejection of a given composite.

To test this hypothesis and to provide some quantitative understanding, six copper specimens with brazed joints of varying quality were made for comparison of their properties with respect to sound transmission. Differences in grain size arising from varying heat treatments have now been eliminated. At present, the temperature-producing system

and the temperature-measuring techniques are being evaluated. It now appears that the specimens may be so long that a masking of the temperature differences from varying bond quality takes place. Efforts are continuing to meet these experimental objections.

ii. Development of improved piezoelectric transducers - Some effort is being put into the development of piezoelectric transducers with improved characteristics compared with those commercially available, for it has been difficult to obtain transducers which have the most desirable electromechanical properties and yet a reasonable underwater lifetime. In addition, there have been serious problems with the uniformity and availability of the commercial products. A simplified mounting method has already been developed, and several successful specimens have been constructed.

b. Neutron Imaging - Further studies of the Juggernaut reactor neutron radiographic beam indicate that the ratio of neutrons/cm² to mr of gamma radiation in the direct beam is of the order of 10⁶. This ratio is sufficient so that direct-exposure techniques can be employed most of the time. Exceptions occur when the object being inspected has high absorption for thermal neutrons and high transmission for gamma radiation. Such a situation was recently encountered in an application involving the inspection of a 4-in. OD-double-walled steel tube having the space between the two walls (about $\frac{1}{8}$ in. wide) filled with boron carbide. The high neutron absorption and relatively large gamma transmission of the object greatly reduced the neutron-to-gamma ratio of the transmitted beam and forced the use of transfer methods in order to obtain a true picture of the neutron transmission of the object. The use of lead and bismuth filters is now being studied in order to determine whether or not a more favorable neutron-gamma ratio can be obtained.

The higher neutron intensity available from the Juggernaut beam as compared with the neutron-imaging facility at CP-5 has made the use of silver and rhodium transfer exposure methods possible. The optimum thicknesses of these materials from the speed standpoint have been determined to be about 0.015 in. for silver and 0.010 in. for rhodium.

c. A Pulsed-field Reflection System for Use with Eddy Current Techniques - In ANL-6610 (Reactor Development Program Progress Report, August, 1962, p. 52), some optimism was expressed in the possibility of improving the signal/noise ratio by substituting a pulsed system for the sinusoidal system in eddy current testing.

On mask-aperture assemblies used on the pulsed-field reflection system, a 0.063-in. aperture has always been standard, but tests on tubing of less than 0.020-in. wall thickness have indicated that an aperture of 0.045 in. might provide a worthwhile increase in surface resolution and S/N ratio. A new mask-aperture assembly was constructed with a 0.045-in.

aperture to check this possibility. This assembly did provide significantly better resolution, but the field strength was down 20 db from the field provided by a mask of identical contour but of 0.063-in. aperture. This loss was great enough to nullify all the expected improvement of S/N ratio. Previous experiments had indicated that the loss resulting from the smaller aperture should not exceed 8 db. Some investigation is necessary in order to understand and correct the factors responsible for the additional 12-db loss.

C. Heat Engineering

1. Heat Transfer between Fluids of Low Prandtl Number in Double-pipe Heat Exchangers

A research effort has been initiated to study heat transfer between nonboiling liquids of low Prandtl number in double pipe heat exchangers. The initial objectives are to develop (1) improved design techniques, and (2) methods of analyzing heat transfer coefficients obtained from such exchangers.

An analytical solution has been obtained for the cocurrent plug flow (i.e., with uniform velocity and thermal conductivity) of two fluids through adjacent, infinitely wide, parallel plane channels separated by a common wall. The plug-flow idealization is known to be a good approximation for fluids with low Prandtl numbers typical of liquid metals and with moderate Peclet numbers. Although application of the analytical solution for this special case is not complete, a particular result of some interest has been obtained, and will be briefly described.

Well-developed Nusselt numbers for the above special case have been found to be a function of three independent dimensionless numbers, H , K , and K_w , defined as follows. Let

c_i = specific heat of fluid "i"

W_i = mass flow rate of fluid "i"

a_i = width of channel through which fluid "i" flows

k_i = thermal conductivity of fluid "i"

k_w = thermal conductivity of common wall

and

b = thickness of common wall.

Also, let subscript "1" identify the fluid for which the Nusselt number is being computed, and subscript "2" identify the fluid flowing cocurrently with fluid "1" and with which heat exchange takes place through the common wall.

Then,

$$H = c_2 W_2 / c_1 W_1 \quad ; \quad K = \left(\frac{a_2}{k_2} \right) \frac{k_1}{a_1}$$

and

$$K_w = \left(\frac{b}{k_w} \right) \left(\frac{k_1}{a_1} \right) .$$

It is found that for $0 \leq K_w \leq 0.1$, the well-developed Nusselt number is not significantly dependent on K_w . Values of K_w within this range are typical of those appropriate to most liquid metal-to-liquid metal heat exchangers. The dependence of the Nusselt number on H and K , however, is found to be of importance. This dependence is shown in Figure 9, in which the well-developed Nusselt number is plotted vs H at constant values of K for the case of $K_w = 0$. Values of well-developed Nusselt numbers for the familiar boundary conditions of uniform heat flux at the wall and uniform wall temperature are shown for comparison. Although the case of uniform heat flux at the wall seems to represent an upper limit for the Nusselt number which is approached as K becomes very large, there is apparently no lower limit.

Figure 9. Fully Developed Nusselt Number for Cocurrent Slug Flow in Adjacent Parallel Plane Channels as a Function of H and K for Case of $K_w = 0$

One possible use of the type of relationship shown in Figure 9 is for the correction of heat transfer coefficients obtained experimentally in double-pipe heat exchangers to values appropriate to a standard boundary condition, such as uniform wall flux. Empirical correlations would then be based on the data corrected to this standard boundary condition. Practical application of the empirical correlation so obtained would then depend on the use of the chart in reverse.

Future work will investigate further applications of the analytical solution for the cocurrent case and the range of validity of the results obtained with the slug flow approximation. An attempt will also be made to obtain an analytical solution for the countercurrent case. It is expected that eventually a set of simple experiments will be performed to support the theoretical calculations.

2. Studies of Boiling Liquid Metals

Initial operation of the forced-convection boiling sodium experiment has been delayed by approximately twelve weeks because of difficulties encountered in the final leak-check in late July.

The leaks in the loop have now been repaired and the maximum leak rate for the entire system was found to be less than 5×10^{-9} std cc/sec. Two differential pressure gauges are being made. Initial operations are expected to begin after the gauges are installed.

Design and cost estimates for the second boiling sodium loop and the higher temperature columbium-1%-zirconium alloy loop are in preparation.

D. Chemical Separations

1. Fluidization and Volatility Separations Processes

a. Fluoride Separations - Additional results have been obtained for the rate of decomposition of gaseous plutonium hexafluoride by alpha radiation. Average decomposition rates for runs of about 21 and 22 days varied from 0.72 to 1.19 percent per day. The runs were carried out in nickel and aluminum vessels having volumes of about 127 cc. Decomposition rates at about 27°C were not appreciably different from the rates obtained at 82°C. In experiments at 82°C, rates obtained with plutonium hexafluoride at an initial pressure of 75 mm Hg were similar to those obtained with an initial pressure of plutonium hexafluoride of 790 mm Hg. A slightly lower decomposition rate was obtained with aluminum spheres than with nickel spheres.

b. Direct Fluorination of Uranium Dioxide Fuel - Fluorination techniques are being developed for the removal of uranium and plutonium from the products of oxidation of uranium dioxide-plutonium dioxide

solid solutions (see Reactor Development Program Progress Report, September 1962, ANL-6619, page 46). The oxidation product is a mixture of U_3O_8 and PuO_2 which contains about 0.3 wt-% plutonium and about one wt-% of a mixture of 10 fission product element oxides. The product of the oxidation was mixed with Alundum before fluorination in all cases except one, in which the Alundum was mixed with the uranium dioxide-plutonium dioxide solution before the oxidation. The latter procedure had no apparent effect on the removal of plutonium from the mixture in subsequent fluorinations. In four experiments, the fluorinations were carried out in two steps. The first step was carried out at 450°C for 2 hr using 10 volume percent fluorine, 25 volume percent oxygen, and 65 volume percent nitrogen; the second step was carried out at 550°C for 5 hr using 75 volume percent fluorine and 25 volume percent oxygen. These fluorinations resulted in the removal of about 99 percent of the plutonium in the original mixture. This is equivalent to a retention of plutonium in the inert solid of about 0.015 wt-%. In one experiment, the two fluorinations were carried out at 500°C, the first using 10 volume percent fluorine, 25 volume percent oxygen, and 65 volume percent nitrogen for one hour, and the second using 100 volume percent fluorine for 5 hr. The results of these fluorinations showed the removal of about 97.2 percent of the plutonium from the original mixture, which is equivalent to the retention of plutonium in the inert solid of about 0.032 wt-%.

These data appear to indicate that plutonium can be satisfactorily removed from alumina by means of a two-step fluorination cycle. In the first step, the uranium is removed by reaction with a low concentration of fluorine, 10 volume percent, at temperatures between 450 and 500°C, and in the second step, the plutonium is removed by reaction with higher concentrations of fluorine, 75 to 100 percent, at temperatures between 500 and 550°C.

Studies of the fluorination of U_3O_8 in the presence of Alundum are being made in a $1\frac{1}{2}$ -in. laboratory-scale fluid-bed reactor. The results of these studies will be used to establish the optimum operating conditions for a fluid-bed apparatus for the fluorination of uranium dioxide-plutonium dioxide mixtures. In the current work, seven fluorination experiments have been carried out at an Alundum-to-uranium ratio of about 2.5 to 1 (the usual ratio is 0.3 to 1). The results have shown that (1) a bed temperature of about 500°C resulted in an efficient, sustained controllable reaction of U_3O_8 with fluorine, (2) the blowback of the filters, to return fines to the reaction zone, is required in order to achieve high conversions of U_3O_8 to uranium hexafluoride, and (3) the dilution of the fluorine inlet stream with nitrogen is necessary for the prevention of caking in the Alundum bed.

Pilot-plant studies have been continued toward the development of a two-zone oxidation-fluorination scheme of uranium dioxide pellets in a single reactor (see Reactor Development Program Progress Report, September 1962, ANL-6619, page 47). In this method, U_3O_8 fines are formed in the lower zone. In this zone, a bed of uranium dioxide pellets is contacted

with an oxygen-nitrogen mixture. Alumina grain is used as a fluidizing medium in the voids of the pellet bed. The uranium oxide fines formed in the lower zone are carried by solids mixing and elutriation into an upper zone of fluidized alumina. Fluorine is introduced at the bottom of the upper zone to form the uranium hexafluoride product. By means of this scheme caking has been prevented and good temperature control at satisfactory fluorination rates has been achieved.

In a recently completed pilot-plant run, a new approach was tested for the purpose of improving operation in the lower zone. The bottom of the reactor was cooled so that the temperature at the top of the lower zone was about 400°C and decreased toward the bottom of the reactor. The upper (fluidization) zone was maintained at about 500°C as in previous runs. Highly satisfactory operation resulted in this run, which used a 12-in.-deep pellet bed charge and a 24-in.-deep fluidized zone above the pellet charge. Oxygen concentrations of 18 and 31 percent in nitrogen were passed through the pellets at a total gas flow of 1.1 cfm (25°C, 1 atm). In the fluorination zone, the total gas flow was 1.3 cfm, and the gas contained 15 percent fluorine. The 12-in. bed of pellets was 91 percent reacted in 13.8 hr with an overall fluorine efficiency of 49 percent. The average production rate of uranium hexafluoride increased from 34 to 44 lb $\text{UF}_6/(\text{hr})(\text{sq ft reactor cross section})$ when the oxygen concentration through the pellets was increased from 18 to 31 percent. However, as the pellet bed decreased, the production rate dropped to 36 lb $\text{UF}_6/(\text{hr})(\text{sq ft})$. This rate probably could be improved by raising the temperature of the bottom of the column as the end of the run is neared. The procedure tested in this run has given improved operation even at relatively high oxygen concentrations. It promises to make reaction control less critical and to allow additional fluorine economy through the use of recycle gas for fluidization.

c. Chlorination of Uranium Dioxide Fuel - If chlorine is used as a reactant in the process for decladding stainless steel-clad uranium dioxide fuels, it is desirable to know to what extent uranium dioxide reacts with chlorine at 600 to 650°C. Two chlorination experiments were carried out. In the first experiment, dry chlorine was reacted with a $1\frac{1}{2}$ -in.-diameter by $\frac{1}{2}$ -in.-long uranium dioxide pellet at 650°C for 3 hr, resulting in a conversion of 48 percent of the uranium dioxide to uranyl chloride (UO_2Cl_2). In the second experiment, the same reaction was carried out with an equal molar mixture of chlorine and nitrogen, at 600°C, resulting in a conversion of 2 percent of the uranium dioxide to uranyl chloride.

d. Separation of Uranium from Zirconium Alloy Fuels

i. Studies of the chlorination and fluorination steps with down-flow fixed-bed filters - The reactant sequence currently being evaluated for the recovery of uranium from highly enriched uranium-zirconium alloy fuels includes hydrogen chloride, phosgene, hydrogen fluoride, and fluorine. In a recent run, the effect of multiple batching during hydrochlorination was

investigated. Three batches of alloy chips were charged during the hydrochlorination step, each charge consisting of about 240 gm of 5 wt-% uranium-zirconium alloy chips ($\frac{1}{8}$ to $\frac{1}{4}$ in. in size). The run cycle time was 28.3 hr, which may be compared with a cycle time of 9 to 12 hr for a single batch of alloy. The run included 21.3 hr of reaction with hydrogen chloride at 400°C (each batch was charged successively and reacted with hydrogen chloride for 4, 4.2, and 13.1 hr, respectively), one hour of reaction with phosgene at 350°C, 2 hr of reaction with hydrogen fluoride at 350°C, 2 hr of reaction with fluorine at 350°C, followed by a second 2-hr fluorination at 500°C. Of the initial charge of uranium, about 0.3 percent passed through the downflow filter during hydrochlorination and phosgene treatment, and about 0.14 percent was retained by the Alundum in the filter bed. A material balance showed that most of the remaining uranium was collected in the cold traps as hexafluoride (98.2 percent). A small amount was removed in sampling (0.7 percent), and about 0.5 percent was collected in the activated-alumina tower for fluorine disposal. Sixty to 80 percent utilization of hydrogen chloride was attained during the major portion of the run. During this period, the hydrogen chloride concentration in the inlet gas was less than 20 percent. Alloy reaction rates of up to 50 gm/hr were obtained while bed temperatures were maintained at the desired levels, about 400°C, without forced cooling. Reaction with hydrogen fluoride, which was expected to reduce the chloride content of the final hexafluoride product, apparently had little effect. Analyses of products obtained from runs made with and without the hydrogen fluoride step showed chloride-to-uranium mole ratios of 0.01 and 0.02.

ii. Fluid bed hydrolysis of zirconium tetrachloride - In order to more nearly simulate conditions proposed for the pilot plant, a feed stream containing a high concentration of hydrogen chloride (about 60 volume percent) was used in the current work, instead of using only nitrogen as the carrier for the tetrachloride. Sand was also re-evaluated as a starting bed material. Five runs ranging in duration from $3\frac{1}{2}$ to $4\frac{3}{4}$ hr were made. In three experiments, zirconium-oxide-coated Alundum was used as the bed material, whereas sand was used in the other two. A hydrogen chloride feed stream was tested once with each material. Zirconium tetrachloride feed rates ranged from about 2.3 to 3.2 kg/hr, and the quantity of steam fed was about three times the stoichiometric requirements.* The bed temperature was 350°C, and the initial bed weight was 20 kg. All of the runs were considered satisfactory in that little pressure drop developed across the filters (80 psig blowback pressure is used) and the quantity of fines that were produced did not appear to interfere with column operation.

e. Conversion of Uranium Hexafluoride to Uranium Dioxide.
Preparation of High-density Particles - The fluid-bed method for preparing high-density, spheroidal, uranium dioxide particles directly from uranium hexafluoride involves the simultaneous reaction of the hexafluoride with steam and hydrogen in the region of 650 to 700°C. Uranium dioxide, with

*Based on the reaction $\text{ZrCl}_4 + 2 \text{H}_2\text{O} \rightarrow \text{ZrO}_2 + 4 \text{HCl}$.

particle densities up to 89 percent of theoretical, had been produced in tests at 700°C. Current work investigated the effects of increased bed height and reduced hydrogen excess on particle density. At a reaction temperature of 650°C, no marked differences were noted when the bed depth was increased from 8 to 18 in. and the hydrogen excess was reduced from 17 to about 4 times the stoichiometric requirement.* The steam rate was 1.15 times the stoichiometric requirement, and the uranium hexafluoride feed rate was 25 gm/min (a 30-min/hr feed time). Density of the dioxide product was about 9.6 gm/cc as compared with 9.3 gm/cc obtained with shallow beds and high hydrogen excesses.

The preparation of larger (up to 14 mesh) uranium dioxide particles for possible application in vibratory-compacted fuel elements was explored in one run. The run was made without addition of seed particles, so that particle growth could occur. A starting bed of +40 mesh material was used. The steam rate was about 1.25 times the stoichiometric requirement and the hydrogen rate was kept relatively low, about 4.2 times the stoichiometric requirement. Nitrogen was used as an additional gas to maintain fluidization requirements. The reaction temperature was 650°C. Particle growth up to 16 mesh occurred during the 15-hr period. Because of the particle growth, it was found necessary to increase the superficial velocity from 1.5 to 3.5 ft/sec to maintain fluidization. The average particle diameter increased from an initial value of 620μ to 704μ during the 15 hr of operation (accumulated over a 3-day period). The final bed of 8 kg contained 2.2 percent +16 mesh particles and 4.0 percent -16 +18 mesh particles, whereas the starting bed was composed of -18 +60 mesh material. The density of the coarse fraction (+18 mesh) was about 8.9 gm/cc (about 81 percent of theoretical). This value is somewhat lower than expected and is attributed to the hexafluoride feed rate which was low at times and which as a consequence, resulted in steam excesses that were high. These conditions give lower particle densities.

Samples of uranium dioxide powder with initial particle densities ranging from 8.5 to 9.75 gm/cc were sintered in hydrogen to achieve further densification. After $2\frac{1}{2}$ hr at temperatures in the range from 1600 to 1725°C, particle densities increased significantly (10.4 to 10.6 gm/cc, equivalent to about 95 to 96.5 percent of theoretical).

f. Fundamental Fluidization Studies - In chemical processes involving gas-solid reactions which are carried out in fluidized beds, the particle size distribution of the solid is an important parameter of the system. Rapid changes in particle size, which may lead to caking or production of fines that may be lost by elutriation, can interfere seriously with successful process operation. A method for the rapid detection of particle size changes is being developed. Experimental fluid-bed apparatus designed to give information on the relationship between gas bubbles in the bed and particle size and particle

*Based on the reaction $\text{UF}_6 + 2 \text{H}_2\text{O} + \text{H}_2 \rightarrow \text{UO}_2 + 6 \text{HF}$.

size distribution has been installed. Experimentation will involve the evaluation of pressure pulses generated by small bubbles of gas admitted to a bed that is maintained at or near fluidizing conditions.

2. Chemical-Metallurgical Process Studies

a. Chemistry of Liquid Metals - The solubility of technetium in liquid zinc over the temperature range from 428 to 758°C may be represented by two empirical equations:

$$(428 \text{ to } 544^\circ\text{C}) \text{ Technetium: } \log (\text{atom percent}) = 6.431 - 7159 T^{-1} ,$$

and

$$(544 \text{ to } 758^\circ\text{C}) \text{ Technetium: } \log (\text{atom percent}) = 9.413 - 15,690 T^{-1} + 4.979 \times 10^6 T^{-2} .$$

The solubilities of the second transition series metals in liquid zinc at 600°C have been measured. The data are summarized in Figure 10.

Figure 10
Solubility of Second Transition Series Metals in
Liquid Zinc at 600°C

The solubility of uranium in liquid zinc may be represented by two equations, one applying above the temperature (~806°C) at which transformation from the epsilon phase to the delta phase occurs, and the other applying in the temperature range from 420 to 806°C:

$$(\text{above } 806^\circ\text{C}) \text{ Uranium: } \log (\text{wt-}\%) = 21.860 - 41,780 T^{-1} + 2.052 \times 10^7 T^{-2} ;$$

$$(420 \text{ to } 806^\circ\text{C}) \text{ Uranium: } \log (\text{wt-}\%) = 7.101 - 6837 T^{-1} .$$

It was found that the ternary solid phase in the uranium-zinc-magnesium system (see Reactor Development Program Progress Report, August, 1962, ANL-6610, page 58) is not formed at any melt composition below 400°C or during the freezing of uranium-magnesium-zinc solutions containing more than 41 percent magnesium at any temperature. It was also found that the composition, 41.0 percent magnesium, 58.9 percent zinc, and 0.1 percent uranium, is a ternary peritectic point in the zinc-magnesium-uranium system. The uranium solubility at 700°C has been found to vary from 4.2 percent in 25 percent magnesium-zinc to about 0.4 percent in 45 percent magnesium-zinc.

The existence of the following intermediate phases in the neodymium-cadmium system was verified by the results of X-ray analysis: NdCd₁₁, NdCd₆, NdCd₄, NdCd₃, NdCd₂, and NdCd. This confirms earlier results obtained by means of the recording effusion balance (see Reactor Development Program Progress Report, August 1962, ANL-6610, page 59).

Measurements of the neodymium-zinc system by the effusion method revealed the existence of the following intermediate phases: NdZn₁₁, NdZn_{8.5}, NdZn₇, NdZn_{4.3}, NdZn_{3.5}, NdZn₃, and NdZn₂. The stoichiometry of these phases was verified by X-ray examination.

b. Liquid Metal Distillation - A sixth run was performed in the investigation of the extent of entrainment of liquid cadmium in cadmium vapor in the large cadmium-distillation unit (see Reactor Development Program Progress Report, August, 1962, ANL-6610, page 59). Study of the results of all six runs revealed that the extent of entrainment, as indicated by the carryover of the nonvolatile solute tin could not be correlated with the rate of evaporation or the power input to the induction coil. Further measurements will be made in which the following factors will be investigated: (1) homogeneity and stability of the cadmium-one percent tin solution in the distillation crucible, (2) representativeness of the distillate samples, and (3) consistency of operating variables from run to run.

c. Uranium Oxide Reactions in Molten Halides - Spectrophotometric studies of uranyl chloride solutions in the NaCl-KCl-MgCl₂ ternary eutectic at 500 to 800°C indicate a thermal decomposition, which has tentatively been ascribed to the reaction

Equilibrium behavior was indicated, since evacuation of the system appears to drive the reaction to the right while the addition of chlorine appears to reverse the reaction.

d. Mass Transfer in Liquid Metals - The influence of agitation on the rate of solution in cadmium of uranium cylinders of 7 mm diameter and 7 mm length was measured for agitator speeds of about 250 to 1400 rpm. As the agitator speed was increased from the minimum, a sharp increase in solution rate was noted at 500 rpm. This indicates that particle suspension began near that agitator speed.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Site and Buildings

An intensive study directed toward final selection of the site location for the AARR facility, such that it best satisfies the needs of experimenters, reactor operations, plant maintenance, and other divisions concerned, has been concluded. Figure 11 is a pictorial view of the AARR facility.

Figure 11. Artist's Rendering of the AARR Facility Made on an Actual Photograph of Selected Site. The View Is towards the West, with the Laboratory and Office Wing of CP-5 (Including the Extension Now Under Construction) Shown at the Left.

The site plan which was finally agreed upon places the AARR facility about 100 ft to the west of the existing CP-5 research reactor. This site has many unique advantages over other plans considered, some of which follow:

- (1) Proximity to CP-5 (Building 330) permits the possibility of a short tunnel connecting the CP-5 and AARR buildings, which would encourage convenient exchange of light equipment, such as instrumentation, between the two facilities.
- (2) Location of the 400-m beam (used for time-of-flight experiments) is such that it interferes least with adjacent property with respect to any future construction in the area.
- (3) Parking facilities can be placed so that AARR traffic is diverted away from already congested areas.

2. Research and Development

a. Core Physics Calculations - The worths of hafnium safety and control rods have been re-evaluated with the use of better cross-section representation and taking into account recent changes in core composition and layout. Hafnium safety rods with zirconium followers are estimated to be worth about 9% $\delta k/k$; six control rods similarly made are estimated to be worth about 13% $\delta k/k$. On a preliminary basis dispersions of Eu_2O_3 in stainless steel appear to be about as effective as hafnium rods.

The response of the reactor to sudden voiding of the central thimble has been given a preliminary analysis. This analysis predicts local boiling but no bulk boiling; if the rods are inserted with an acceleration of 2.5 g after a 50-ms delay, the hottest plate does not go over 650°C. These are modest requirements and the detailed analysis should not change them by large factors; a conventional control system therefore appears adequate.

b. Critical Experiment - All design work on cell modifications for the AARR critical experiment has been completed. Construction is scheduled to begin immediately after drawings are checked.

The predicted critical mass of the clean critical experiment is 13 to 14 kg of U^{235} , depending on the amount of hydrogen in the outer reflector. The error allowance is 1 kg for 2% δk .

Reflector worth vs 100% water is about 7% for heavily loaded cores, more for lightly loaded cores.

c. Preliminary Hazards Analysis - A study has been made of plume dosage under extreme conditions: primary system rupture and complete core meltdown after 12,000 Mwd. The plume dose arises from the leakage rate tolerance established for the containment shell, namely, 0.1% per day. The study showed that the exclusion radius is determined by the concentrated source in the field building, not by the plume dosage, and is 235 m. The low-population zone radius extends to 3.5 km under the worst atmospheric conditions. This requirement is met at the AARR site.

Further study of the plume dose considered the effects of continued ventilation after the incident, as opposed to sealing the building. If, for example, the building is ventilated at 6000 cfm (similar to practice at the Oak Ridge Research Reactor), with filtration for particulate matter and halogens, the exclusion radius and low-population zone extend only to the containment shell. The filtration equipment is located underground in a shielded underground vault. Under these conditions, emergency work in the neighborhood of the reactor could begin shortly after a possible incident, and work at nearby Laboratory installations would suffer only minor interruption.

d. Core Heat Transfer and Hydraulics - A heat transfer study of proposed AARR cores was completed to determine the minimum core inlet pressure as a function of coolant velocity for various power levels.

Two 77-liter AARR flux trap cores were compared. One utilized 0.040-in. (0.1-cm) fuel plates and channel spacings (Case 1), whereas the other (Case 2) used 0.050-in. (0.13-cm) fuel plates and 0.030-in. (0.076-cm) channel spacings. The core lengths were both 18 in. (45.7 cm), and identical max/avg power ratios, hot spot factors, and power distributions were assumed. The designs were limited by the criterion that local boiling would be incipient at the surface hot spot. The study also determined the power increase margin that would cause instability for parallel-channel flow, which was assumed to occur with the initiation of net steam generation at the exit of the hot channel.

Although physics calculations have indicated higher neutron fluxes for the Case 2 core (see Reactor Development Program Progress Report, August 1962, p. 62), the heat transfer study showed that the Case 1 core requires about 30% less pumping power and has a higher margin of safety (1.6 as compared with 1.35) against flow instability for the same system pressure. The margin against film blanketing burnout is identical for both cases. The study, which was limited to the heat transfer and fluid flow aspects of the cores, also showed that, although a higher coolant velocity might be expected to yield a lower system pressure, there is a point at which the frictional pressure drop is so large that a higher system pressure would be required even with increased velocity. Coincident with this phenomenon is a reduction in the power increase margin against flow instability as the velocity is increased.

The determination of the flow instability margin on the basis that net steam generation at the channel exit causes flow instability is recognized as merely a convenient calculational tool, but comparisons made between the two cases are nonetheless valid. The study pointed out that the highest core performance can be achieved when the film blanketing burnout margin and the flow instability margin are the same. In the cases studied it was assumed that all reactor channels were identical, but it is clear that the core could be designed to provide more sophistication by utilizing such devices as inlet orifices in the channels, roughening of channels, variable channel spaces, and side venting of channels, to increase the margin against flow instability. In the cases studied, however, it was concluded that Case 1 involved less complexity and was therefore better from a heat transfer and fluid flow viewpoint. The relative value of the additional attainable neutron flux in Case 2 in terms of the additional complexity and reduced power increase margin, remains to be evaluated.

B. Undersea Application of Nuclear Power

A parametric study is underway to find the maximum power/weight ratio for small, highly enriched, pressurized water systems to be used for undersea applications. A schematic diagram of a typical pressurized water system is shown in Figure 12. The purpose of the parameter study is to determine the optimum combination of design specifications and operating conditions which give the maximum power/weight ratio. Simplified models for the calculation of reactor physics, shielding, and heat transfer are being developed to keep the study within reasonable proportions. These models will be accurate enough to show the interrelation of the components and to point out the most promising areas for more detailed design studies in the small sizes desired. Parameter studies will be made for other systems, such as boiling water and liquid metal-cooled reactors.

Figure 12

The Layout for a Pressurized Water Reactor of the Type Considered for Undersea Power Applications.

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal Oxidation and Ignition Studies

Further calculations of the course of uranium ignitions have been completed. In these studies, rate equations for the uranium-oxygen reaction (see Reactor Development Program Progress Report, June 1962, ANL-6580, page 52) are used in conjunction with a simple heat transfer model. A step-wise calculation method is used to predict the course of burning-curve ignition experiments. Previous results indicated excellent agreement between theory and experiment for specimens having specific areas of 0.5 and 5 sq cm/gm. Recent computations have been concentrated on finding the conditions which, for a small change in specific area, would allow the transition to a protective oxide to occur at 450°C and thereby inhibit immediate ignition. Calculations indicated that specimens having a specific area of 5 sq cm/gm or less had an ignition temperature of 510°C or higher, whereas specimens having specific areas of 7 sq cm/gm or greater ignited at 385°C. Experimentally, the break occurs in the range 2 to 5 sq cm/gm. The greater tendency, found experimentally, to ignite is believed to be caused by a change in actual surface area during the oxidation which precedes the ignition.

Burning-curve ignition studies of uranium alloys described previously (see Reactor Development Program Progress Report, August 1962, ANL-6610, page 65) were re-examined. It was concluded that there are only two major factors operating to alter the shape of the unalloyed uranium-burning curves, although they may occur in combination. One of these effects, shown particularly by aluminum, was to inhibit the transition to a protective oxide above 450°C. A number of other alloying agents caused rapid self-heating below 450°C, and it might be presumed that these agents also inhibit the transition. The second effect, which is important to the understanding of uranium alloy ignitions, is the tendency of certain alloying agents to render the high-temperature oxide more protective. This effect was noted particularly with copper alloys.

Measurements have been made of maximum burning temperatures and burning-propagation velocities for several zirconium foils burning in pure oxygen. These results, which were obtained with the aid of a two-slit optical pyrometer, are consistent with unpublished data obtained by means of high-speed cinematography. Burning temperatures up to 3000°C were observed.

Helium-shielded ignition studies of various sizes of spherical zirconium powder have been completed with -140 +170 and -325 +400 mesh powder samples ranging from 0.01 to 3.0 gm in $\frac{1}{4}$ -, $\frac{3}{8}$ -, and $\frac{1}{2}$ -in. crucibles. The following relationship between modified sample depth and ignition temperature $T(^{\circ}\text{K})$ is obtained:

$$1/\text{depth} = K e^{-12050/RT} .$$

Modified sample depth was computed as follows:

$$1/\text{depth} = (1/d) + (1/h) \quad ,$$

where d is crucible diameter (cm), and h is depth of sample (cm). Values of K for the two powders studied were as follows:

-140 + 170 mesh powder ($105\text{--}88 \mu$ diam) $K = 2.3 \times 10^4/\text{cm}$;

-325 + 400 mesh powder ($44\text{--}35 \mu$ diam) $K = 7.2 \times 10^4/\text{cm}$.

Isothermal oxidations of plutonium, over the temperature range from 140 to 320°C, indicate an initial parabolic oxidation process with an activation energy of 32.7 kcal followed by an apparent linear rate.

2. Metal-Water Studies

Studies of the aluminum-water reaction by the levitation melting method are continuing. The experiment consists of suspending a ball of aluminum in an atmosphere of steam by means of a radiofrequency field. The aluminum is then rapidly heated to the test temperature at which the sample is maintained for a period of time. The amount of reaction is obtained by a gravimetric determination of the aluminum oxide (Al_2O_3) produced. Preliminary studies at 1400, 1500, and 1600°C have shown that the reaction is linear. Typical values for 10-min reaction times are 1.28 ± 0.33 , 3.19 ± 0.59 , and 7.7 ± 0.2 mg/sq cm aluminum obtained at 1400, 1500, and 1600°C, respectively.

In-pile studies of metal-water reactions in TREAT are continuing. A series of eight experiments with aluminum-uranium alloy plates were conducted. Preliminary results of five experiments with 60-mil-thick, 0.5-in. by 1.35-in. plates containing a 20-mil-thick core of fully enriched 23 weight percent uranium-aluminum alloy showed progressively greater damage with increasing reactor power. Metal-water reaction ranged between 0.10 and 10.4 percent for reactor power ranging from 170 to 500 megawatt-seconds.

B. Fast Reactor Safety Studies

1. Meltdown Experiments

Experiments are being performed with TREAT to study the characteristics and causes of failure of fast reactor fuel elements, and to survey the relevant mechanisms influencing fuel movement.

a. High-temperature Transient Behavior of Uranium Oxide Specimens - Post-excursion inspections were completed on the four 0.381-cm-diameter uranium oxide fuel samples of Series XXXIII.

Each specimen was clad with a refractory metal, EBR-II-sized jacket and thermally bonded with argon gas. Samples Nos. 1 and 2 were clad with niobium, and samples Nos. 3 and 4 with tantalum. The maximum cladding temperatures recorded during the excursions were 2180°C, 2300°C, 2300°C, and 2480°C, respectively, for the four elements. Elements Nos. 3 and 4 were each fitted with an internal tantalum-sheathed-tungsten-5% rhenium versus tungsten-26% rhenium thermocouple. Both internal thermocouples indicated fuel melting occurred, but the actual maximum internal temperatures are not known because the output was well above the temperature limit of 2320°C for which the thermocouples had been calibrated by the manufacturer.

The claddings of elements Nos. 1, 2, and 3 were warped, but did not fail. The bottom 2 cm of samples Nos. 1 and 2 (the niobium-clad samples) were covered with a very thin layer of scale.

High-speed color motion pictures made during the high-temperature transient of element No. 4 showed a general and extensive failure along the top half of the element. The fuel fragments which were ejected were appreciably smaller than the (unfailed) cladding diameter. The dimensions of the fragments could not be measured because the high-temperature thermal glow made the sample appear larger on the film than the actual size. The expulsion of fuel occurred without violence. The inspection of the No. 4 capsule showed that fuel ejection from the cladding was complete, except for one piece about 1 cm long.

There was no evidence of the oxide cylinders fusing to the cladding in Samples Nos. 1, 2, or 3. Sections of cladding recovered from element No. 4 were metallographically examined. Large crystal grain boundaries characteristic of high-temperature exposure were observed, and some evidences of melting were found on the inside of the clad surface. However, nothing was noted that could be identified as a tantalum-uranium oxide diffusion reaction or interaction zone. The tips of both internal thermocouple sheaths were melted away (that of No. 4 being much worse than No. 3). There was also no clear evidence of a metal-oxide reaction on the sheaths.

The appearances of the individual fuel cylinders from the unfailed samples were similar to those found in other specimens reaching temperatures above 1600°C. The amount of the fine oxide particles present was not appreciable, nor was extensive radial or circumferential cracking observed. However, the cylinders, which were nominally about 3.4 cm long before exposure, were found to be fractured into segments approximately 0.8 cm long. The fragments recovered from the No. 4 capsule were small; about 35% by weight of those collected passed through a screen sized for 0.83-mm-diameter particles.

b. Study of Uranium Sulfide - About 250 linear cm of 0.381-cm-diameter uranium sulfide pellets having a sintered density of about 91% theoretical are being prepared for transient exposure in TREAT. The uranium enrichment is 10%, so that energy input data for the sample can be compared directly with those obtained with the UO_2 specimens. The theoretical density and heat of vaporization of the sulfide are similar to those of uranium dioxide, but the vapor pressure of the sulfide is about one-sixth of that of UO_2 in the temperature range from 2500 to 2750°C. Its behavior under thermal stress may be appreciably different from that of UO_2 .

2. Integral Sodium Loop Development for Experimental Meltdown Studies

A small, forced-convection sodium loop is being developed for use in TREAT to permit the transient meltdown of one to seven EBR-II, Mark-I-sized samples in flowing sodium. The loop fits as an integral unit into the 10 cm square by 240 cm long volume of a TREAT fuel element. Hence, the loop can be manipulated by the regular TREAT fuel-handling equipment. Connections for instrumentation and power to the loop heaters and electromagnetic pump are provided by means of a special top fitting. The entire assembly fits into a stainless steel TREAT fuel can. Figure 13 is a view of a partially disassembled loop showing, from right to left:

(1) The fuel-element support tube fixture which holds the sample element (or elements) in the test section. It also defines a flow channel for upward flowing sodium around the specimen. The specimen is located at the bottom of the support tube and extends upward about one third of its length.

(2) A section of the assembled loop, top to bottom, the electrical connections, sodium reservoir, clam shell heater, electromagnetic sodium pump, and pump transformer.

Figure 13.

Partially Disassembled Integral Sodium Loop. On the Left Is the Steel, TREAT Fuel Can, with Top Fitting and Plug for Electrical Connection above the Can. In the Center Is the Loop Proper. On the Right Is the Sample Fuel-Element Support Tube Fixture.

(3) The steel TREAT fuel can. This can also provides a gas-tight seal for the loop and permits an inert cover gas blanket to be maintained.

The testing sequence is as follows. The loop is assembled and sealed at the DuPage site and is sent to TREAT at Idaho in a special shipping coffin. At TREAT it is transferred to the regular TREAT fuel-handling cask which is used to insert the loop into the reactor core. After loading into the reactor, the electrical and instrumentation connections are made by plugging the leads into the top fitting. The loop is then heated to the specified temperature, sodium flow is initiated, and the reactor power transient is performed to complete the experiment. After the transient, the pump is stopped. The loop is cooled to room temperature, disconnected, removed from the reactor, and shipped back to the DuPage site for disassembly.

Preparations are underway to perform calibration runs in TREAT to establish the correlation between sample power and reactor power. Four loops have been constructed and are ready for installation of pressure transducer standoff subassemblies. The loop shipping coffin which is designed to handle either normal or irradiated specimens is now under consideration.

3. Operation and Maintenance of TREAT

No transient experiments were run during the month because of difficulties encountered in the fabrication of fuel samples. This shutdown time was utilized for maintenance work on reactor equipment and instrumentation.

The shielding support and seal assembly for the sodium loop were installed in the pipe opening between the sodium equipment room and the subpile room. This assembly will support the shielding required between the sodium pipes and the subpile room, and prevent sodium smoke from entering the subpile room through the pipe opening in case a leak in the sodium system causes a fire. The rectifier for the electromagnetic pump for the sodium loop was installed in the sodium equipment room. Assembly and installation of electrical control equipment and instrumentation for the loop was continued during this period.

VI. PUBLICATIONS

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