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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

September 1963

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Issued October 15, 1963

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:

March 1963	ANL-6705
April 1963	ANL-6717
May 1963	ANL-6739
June 1963	ANL-6749
July 1963	ANL-6764
August 1963	ANL-6780

TABLE OF CONTENTS

	<u>Page</u>
III. General Reactor Technology	31
A. Theoretical and Applied Nuclear Physics	31
1. Elastic Neutron Scattering	31
2. Ultra-low-energy Proton-recoil Measurements	31
3. High Conversion Critical Experiments	33
4. Theoretical Physics	34
B. Reactor Fuels Development	38
1. Corrosion Studies	38
2. Ceramic Fuels	39
3. Particulate Metal Fuel Elements	42
4. Extrusion Developments	46
5. Nondestructive Testing	46
C. The Metallurgy Alpha-Gamma Hot Cell	48
D. Heat Engineering	49
1. Refractory Metal Test Loop	49
2. Boiling Sodium Two-phase Flow Studies	49
E. Chemical Separations	49
1. Chemistry of Liquid Metals	49
2. Fluidization and Volatility Separations Processes	50
3. General Chemistry and Chemical Engineering	54
4. Calorimetry	55
F. Plutonium Recycle Program	55
1. Hydrodynamics	55
2. Plutonium Recycle Fuel	56
3. Plutonium Recycle Control Rods	57
IV. Advanced Systems Research and Development	59
A. Argonne Advanced Research Reactor (AARR)	59
1. Facility Design	59
2. Core Physics	59
3. Critical Experiment	60
4. Studies of Reactor Control	60
5. Fuel and Core Design	60

TABLE OF CONTENTS

	<u>Page</u>
6. Design of Experimental Facility Beam Tube	61
7. Shielding	61
B. Magnetohydrodynamics	61
V. Nuclear Safety	63
A. Thermal Reactor Safety Studies	63
1. Metal Oxidation and Ignition Studies	63
2. Metal-Water Studies	65
B. Fast Reactor Safety Studies	65
1. In-pile Experiments of Pre-irradiated Metallic Elements	65
2. Analysis for Coherence of Failure in Reactor	66
3. Analysis of Pressure Surges in Fast Reactor Coolant Channels	68
C. TREAT	69
1. Small TREAT Loop	69
2. Large TREAT Loop	69
3. Experiments in Progress	70
VI. Publications	71

I. BOILING WATER REACTORS

A. BORAX-V

1 Operations and Maintenance

Preparations are proceeding for power operation with central superheater core CSH-1. Superheater fuel assemblies have been repaired by welding and superheater fuel and in-core instruments have been loaded.

Small leaks in the superheater fuel assemblies were detected and repaired prior to operation at zero power (at 489°F and 600 psig). These leaks were in weld seams where the five fuel elements in an assembly are joined to top and bottom flow vanes which maintain the moderator gap spacing between elements at this joining point. To improve the integrity of this joint, a fillet weld was made on every joint in every assembly. This required 16 seam welds per assembly in slots 0.365 in. wide and 3.875 in. long. A special welding machine, which shielded the radioactive fuel assemblies (0.5 to 2 r/hr on contact) and allowed viewing and remote manipulation of the inert gas-shielded tungsten electrode within the slot, was developed for this purpose.

Measurements of the superheater fuel assemblies before and after welding indicated only a slight shrinkage in width and breadth, so that the dimensions of the moderator water gaps are essentially unchanged. Shrinkage in overall assembly length averaged about 0.050 in., which should result in a downward superheater fuel displacement of half that amount. All the assemblies required straightening. All assemblies were final leak-tested by filling with water, and any leaks that still existed were repaired.

While repairing two of the instrumented superheater fuel assemblies, the bottom steam thermocouples were damaged and replaced. New thermocouples were placed in the lower nozzle, bringing the extension leads through stainless steel tubing mounted external to the assembly.

After repair, ten standard superheater fuel assemblies and two instrumented superheater fuel assemblies (one inlet and one outlet) were loaded into the core. The loading diagram for fuel and in-core instrumentation for core CSH-1 is shown in Figure 1. In addition to the instrumented superheater fuel assemblies containing fuel and steam thermocouples and a steam flowmeter venturi, the following in-reactor vessel instrumentation and associated support brackets have been loaded in locations shown in Figure 1:

- a. one instrumented boiling fuel assembly, containing fuel and coolant thermocouples and entrance and exit turbine-type flowmeters;

Figure 1. Central Superheater Core CSH-1 for BORAX-V

b a group of 12 thermocouples for sampling core inlet and exit water temperatures. Two thermocouples are provided at each of the three inlet and exit locations;

c. six $\frac{5}{32}$ -in.-dia stainless steel pressurized flux wire thimbles, three in superheater moderator gaps and three in boiler coolant channels;

d five $\frac{3}{8}$ -in.-dia stainless steel pressurized miniature ionization chamber thimbles replacing flow rods in the boiler.

e. two individual boiling thermocouple fuel rods. As well as furnishing fuel temperature data during operation, these rods will be used later to measure the effect of irradiation on the thermal time response of boiler fuel. The thermal response time of these unirradiated rods has been measured (see Section 2a);

f. a magnetic drag disc-type flowmeter in the downcomer;

g a fuel-poison oscillator rod located in a reflector thimble.

The in-vessel demister-type steam dryer was also placed over the risers to the six inlet superheater fuel assemblies, and a new saturated steam collector was fabricated and installed. The reactor vessel head has been installed, along with numerous packing glands and seals for in-vessel instrument leads on the head nozzles. The remotely controlled steam sampling probe and the acoustic reactor water-level probe were installed on head nozzles. The connection of leads and checkout of in-vessel instrumentation are progressing.

The relocation of the water-control system for the control rod drive seal to the main floor was completed. The presence of the controls on the operating floor rather than in the subreactor room permits manual adjustment and observation during power operation of the reactor.

Installation of the new low-rate superheater flood valve was completed. This valve and its associated manual valve will be adjusted so that a normal flooding time of 10 min is obtained.

2 Development and Testing

a Determination of the Boiling Fuel Rod Time Constant. The contribution of the thermocouple response time to the total thermal response time of BORAX-V instrumented boiling fuel rods has been measured. This $\frac{3}{8}$ -in.-dia fuel rod is made of 4.95 w/o enriched UO_2 pellets clad with 0.015 in of Type 304 stainless steel and has a tantalum-sheathed, BeO-insulated, W/W-26% Re thermocouple spring loaded into a blind hole along the axis of the fuel.

Data for thermocouple time response were obtained by nuclear heating of the fuel rods during a fast transient in the TREAT reactor. The

rods were individually irradiated for 34 Mw-sec, which gave a temperature rise of about 400°C from room temperature. The steel capsule containing a fuel rod during the irradiation was evacuated and the fuel rod wrapped in aluminum foil to reduce heat losses to a minimum.

The thermocouple time response and total instrumented fuel rod time response, measured out-of-reactor by thermal cycling in NaK,¹ for the three instrumented fuel rods tested are given in Table I.

Table I. 63% Thermal Time Response (Time Constant τ) of Instrumented Boiling Fuel Rods for BORAX-V

<u>Fuel Rod</u>	<u>τ of Total Rod (sec)</u>	<u>τ of Thermocouple (sec)</u>
VA-1	6.7	0.98
VA-2	8.0	0.90
WA-8	9.7	1.50
Average	8.1	1.12

b. High-temperature Thermocouples. Two experimental W-W/26% Re thermocouples, with W/26% Re sheaths and high-purity thoria (swaged construction), have been tested to about 5350°F. The tips of both units melted during the last of two cycles. Output of the two couples is shown in Figure 2, with all data from the four furnace cycles (two cycles of which were made in a new installation) shown. The standard curve for W-W/26% Re fails considerably above the curve obtained from the test. It is possible that the required hot-working process used for this type of construction has produced considerable change in the thermocouple wire, a phenomenon easily experienced on noble metal and base metal couples. The samples are now undergoing metallurgical examination to uncover further evidence.

A contract has been awarded to a commercial testing agency for completion of a series of tests (with a 5050°F upper limit established) of six new sample couples, with sintered thoria insulation lightly swaged into W-W/26% Re tubing, and with W/5% Re-W/26% Re conductors.

c. Acoustic Reactor Water-level Probe. The drive for the acoustic water-level probe was completed, and the probe drive assembly was installed for test in the 600-psi autoclave. The probe operates on the following principle: A 28-kc driving sound signal is sent down the probe shaft to a rectangular plate. The mechanical impedance of the water or steam changes the phase of the reflected signal returned up the shaft. Two pickup crystals

¹Reactor Development Program Progress Report, August 1963, ANL-6780, pp 4-5.

Figure 2. Calibration of Two W/W-26% Re Thermocouples in Argon and Helium Atmospheres

at the top of the shaft pick up both signals, and the phase relationship of the driving and returned signals is measured and converted to an electrical signal.

The unit was first hydrotested at 680 psi, cold, and the drive operated to insure adequate operation. Tests were run by keeping the autoclave water level constant and visible in bull's-eye sight glasses, and

Table II. Calibration of Acoustic Water-level Probe

Temperature (°F)	Meter Output	
	In Vapor	In Water
75	0.4	0.9
200	0.51	0.84*
490	0.26	0.84 0.6**

*Nonboiling.

**Boiling. Two-phase mixture at probe.

running the probe up and down out of contact and in contact with the water surface. Meter readout (confirmed by the sight glass) gave immediate evidence of water, water-steam mixture, or steam immersion of the probe, with very uniform performance over a 2-day interval and with temperatures ranging from 75 to 489°F. Meter readout is calibrated from "0" to "1," with "0" representing the probe's immersion in a low-density region and "1" representing a high-density region. Typical readings for various conditions are shown in Table II. Note that the range of readings shown under "Output in Water" illustrates the sensitivity of

the probe to steam void fraction, a factor which is being considered for use in direct void fraction measurement.

3. Superheat Fuel Elements

a. Type 406 Stainless Steel-clad Superheat Fuel. Fifty Type 406 stainless steel-clad enriched-fuel plates fabricated by Atomics International have been received and evaluated. Three were rejected because of inadequate side cladding and were returned to the vendor. The 47 acceptable plates are being descaled with sodium hydride.

Development of subassembly brazing techniques is progressing. A dummy four-plate subassembly was vacuum-furnace brazed with Coast Metals 60 alloy at 1160°C. The surfaces of the dummy fuel plates had been prepared for brazing by (1) removing surface oxide (sodium hydride descaling) and (2) applying a 5×10^{-3} -mm (2×10^{-4} -in.) nickel electroplate to the braze-affected areas. (The necessity for plating was outlined in the January Progress Report, ANL-6683, p. 6.) The brazed joints attained were entirely satisfactory. As expected, the grain size in the stainless steel was large after brazing.

Premabraz 128 (a Au-Ni-Cr alloy) has been used successfully to braze small nickel-plated Type 406 stainless steel specimens. Its flowpoint, originally reported to be 1000°C, is actually closer to 1035°C. At 1035°C

grain growth in Type 406 stainless steel is just as extensive as at 1160°C (the Coast Metals 60 brazing temperature).

Specimens brazed with Nioro (a Au-Ni alloy) at 990°C did not exhibit extensive grain growth. Nioro penetrates more deeply into the base-metal grain boundaries than the other two alloys. A specimen has been submitted for testing in oxygenated steam at 650°C and 600 psi. If results of this testing are satisfactory, a four-plate dummy-fuel subassembly will be brazed with Nioro for evaluation.

b. Advanced Superheat Fuel Elements.

(i) Fabrication Development The Torrington Company, Torrington, Conn., is developing swaging methods for forming mockup pea-in-a-pod-type fuel rods (June Report - ANL-6749, p. 4). The first two samples produced were recently received at ANL for evaluation. There were no folds or wrinkles in the 406 stainless steel tubing between the balls, as had been produced by cold isostatic pressing (see Progress Report for May 1963, ANL-6739, p. 4). A metallographic examination of sections from one of the swaged specimens revealed no cracks.

To be acceptable, the swaging method requires refinement in three respects.

1) The tubing at the rod-diameter minima was reduced to approximately 0.625 cm (0.250 in.) O.D. A reduction to 0.430 cm (0.172 in.) is desired.

2) The tubing wall had been thinned at the rod-diameter maxima from 0.25 mm (0.010 in.) to approximately 0.13 mm (0.005 in.).

3) The balls inside the tube were not touching and were not uniformly spaced apart.

It is likely that a reduction between balls to 0.430 cm (0.172 in.) O.D. is not possible by swaging without thinning the tube wall or pushing the balls apart. The Torrington Company has agreed to send additional specimens which have been swaged as close as possible to the desired dimensions. These specimens will then be isostatically cold pressed at ANL in an attempt to achieve the final size without wrinkling the tubing between balls.

(ii) Alloy Development. Incoloy 800 and Type 406 stainless steel are presently being considered as cladding material for another advanced superheat fuel design: the spherical-pellet concept. Forming and welding of Incoloy 800 have been accomplished without serious difficulties. Forming of 406 stainless steel has also been done successfully; however, all weldments of 406 stainless steel have been brittle. Grain growth in the heat-affected zones has, in every case, been extensive.

A series of six arc-cast alloys are being corrosion tested and evaluated for possible use as superheat fuel cladding. Weight losses after one week exposure to oxygenated steam at 650°C and 600 psi are shown in Table III. The increase in corrosion resistance with increasing aluminum content is clearly evident.

Table III. Corrosion in Oxygenated Steam of Alloys Based on Types 406 and 405 Stainless Steel

Type of Stainless Steel and Nominal Aluminum Content (w/o)	Addition (w/o)	Weight Loss (mg/cm ²)
406; (3.25-4.50 Al)	None	4.3
406; "	1 Ti + 1 Y	4.2
405; (0.1-0.3 Al)	1 Ti + 1 Y + 3 Al	6.3
405; "	1 Ti + 1 Y + 2 Al	10.7
405; "	1 Ti + 1 Y + 1 Al	16.6
405; "	1 Ti + 1 Y	21.7

Some of the arc-cast Type 406 stainless steel with the titanium-plus-yttrium addition was rolled to 0.25 mm (0.010 in.) and annealed to produce an equiaxed grain structure. Preliminary weld studies of this material have indicated that the titanium-and-yttrium addition restricts the catastrophic grain growth usually observed in Type 406 stainless steel following a normal welding operation.

II. LIQUID-METAL-COOLED REACTORS

A. General Fast Reactor Physics

1. ZPR-III

The past month has been largely occupied with preparations of ZPR-III plutonium operations. The plutonium inventory is in the form of small pieces individually canned in stainless steel. Although the canning is quite carefully welded and gives every indication of being sturdy and satisfactory, considerable effort has been expended in preparing to monitor for leaking plutonium in case of a break in the canning. Plutonium is ordinarily detected by its alpha radiation and, in the case of ZPR-III, the ordinary problems of alpha monitoring are further complicated by the fact that a low level of alpha contamination is already present as a result of a number of years of operation with uranium. An extensive cleanup program for the assembly room equipment and inert materials was therefore undertaken to reduce the level of uranium contamination. The enriched uranium fuel was cleaned and re-coated with a Kel-F lacquer, in order to provide color identification and to reduce the spread of alpha activity. All stainless steel pieces which might have become confused with the plutonium pieces were marked with an identifying color. Floors were cleaned and working tables re-covered for a clean start with plutonium. A restricted area, in which protective clothing is required, has been marked off in order to prevent the spread of contamination throughout the building.

Alpha-monitoring equipment has been installed in the reactor room and loading room. An instrument is also available for measurement of neutron dosage arising from the spontaneous fission of the Pu^{240} contained in the fuel.

Combustible material in the reactor room has been reduced by storing the sodium in a separate building. The exhaust ventilation filters have been replaced by a more temperature-resistant type, so that in the event of a fire they would withstand any credible rise of air temperature.

The reactor assembly itself has a new low-speed motor installed on the carriage drive, reducing the final speed of approach to a third of its previous value. This permits the assembly of plutonium criticals with no greater rate of reactivity addition than for uranium criticals, even though the delayed-neutron fraction for plutonium is considerably smaller. Thermocouples were installed in the core region to provide for axial and radial temperature traverses, since the alpha decay of plutonium generates significant quantities of heat.

All tubes in the ZPR-III electronic circuits have been checked, and new BF_3 proportional counters installed.

The first plutonium assembly will be a mockup, simplified for physics studies, of the Rapsodie reactor. The blanket has been constructed, and the control rods placed in the core region. Operation of the rods was rechecked, and core assembly was started. About 27 liters of an estimated 36-liter core have been loaded.

2. ZPR-VI

The ZPR-VI facility went critical for the first time in July 1963 with an all-metal assembly having a U^{238}/U^{235} atomic ratio of about 7. The core height is 50.8 cm and the diameter is 55 cm. The core is surrounded by a 30-cm-thick high-density uranium blanket. The volume fractions for the composition of Assembly No. 1 are as follows:

<u>Composition</u>	<u>Core</u>	<u>Blanket</u>
U^{235}	9.51	0.18
U^{238}	70.8	82.7
Stainless Steel	10.8	10.8
Voids	8.89	6.32

The core composition is about the same as that of Assemblies 11 and 22 studied with ZPR-III. Assembly 1 of ZPR-VI has an additional 1.6 v/o steel in the core and an additional 2.5 v/o steel in the blanket. The critical mass for Assembly 1 has been found to be 234.8 kg U^{235} as compared with 240.6 kg for Assembly 11 on ZPR-III. The lower critical mass is thought to be due to the additional steel in the blanket.

Measurement of the U^{238}/U^{235} fission ratio about 5 cm from the center of the core by means of a thin-walled Kirn-type gas-flow counter gave a value of 0.0392 ± 0.0005 , which may be compared with a value of 0.036 obtained for Assembly 11.

Prompt-neutron period measurements (Rossi-alpha) resulted in a value of $(10.8 \pm 0.7) \times 10^4 \text{ sec}^{-1}$ for β/ℓ . Measurements with Assembly 11 yielded a value of $(10.4 \pm 0.3) \times 10^4 \text{ sec}^{-1}$.

Power calibration, gap-worth experiments, reproducibility tests, central reactivity coefficient measurements of various samples, and control rod calibration studies have also been made with the first assembly on ZPR-VI. The results from these measurements are currently being analyzed.

The training of the staff in the operation of ZPR-VI has continued.

3. ZPR-IX

The ZPR-IX facility will be used initially for a series of critical experiments in connection with the nuclear rocket reactor program. Increased attention is being given to the assembly and testing of this facility as personnel complete similar responsibilities for the ZPR-VI system.

The electrical preparations required for startup are essentially complete. Some difficulties are being experienced with the period channels (as also were experienced with ZPR-VI).

Delays have been encountered in the procurement of acceptable mechanical components. The aluminum matrix tubes have been welded into bundles, and a number have been delivered. A number of bundles still require holes to be drilled for the flux traverse tubes. The drawers have all been delivered. Studies of the hydrogen content of proposed hard-anodized coatings have been completed. The safety rod drives need reworking because of a weakness in the fastenings of the drive rods. The loading conveyor has not been delivered.

All of the bundles to be electrofilmed should be drilled and ready for shipment by the end of this month. All others not requiring electrofilming will be finished later.

Inasmuch as the excessive hydrogen (~100 ppm) present in the "Sanford" coating is inherent, a new approach was attempted. Four sample drawers were sent to Industrial Platers, Inc., in Columbus, Ohio, for plating with a 0.0003- to 0.0005-in. thickness of "X-O-Nickel." The platers claimed that drawers coated in this manner should be equal to the stainless drawers in surface hardness, and should resist corrosion and guard against metal pickup and galling.

The sample drawers were tested thoroughly and analyzed chemically. The "X-O-Nickel" coating proved to be much too soft to provide a good wear surface. Consequently, the "Sanford" hard anodized coating will be used in spite of its high hydrogen content. 90% of the total lot of 15- and 24-in. drawers will be coated. 10% will not be coated, so that the effect of hydrogen in the core can be determined by substitution experiments. All of the 23-in. blanket drawers will be coated. Requests for price quotations have been initiated, and drawings are being revised so that the job may proceed as quickly as possible.

The design of the detector system for flux mapping in ZPR-IX is complete. This design employs four solid-state detectors situated in a square block of aluminum that will be driven remotely by a step motor in a way that provides precise positioning. The four detectors will be covered by materials such as B^{10} , U^{235} , Pu^{239} , and U^{238} , and will be used to detect

fission fragments and alpha particles. The detectors will be connected to the preamplifiers (about 2 m away) by enameled wires in order to avoid the introduction of hydrogenous components of conventional insulation in the core. It is not certain how the capacitive coupling between the enameled wires will affect the output of each detector. An experiment to determine this effect is underway.

B. General Fast Reactor Fuel Development

1. Fast Reactor Fuel Jacket Development

a. Finished Tubing for Irradiation Tests. Small quantities of Nb-10 w/o W-1 w/o Zr-0.1 w/c C (D-43), Ta-8 w/o W-2 w/o Hf (T-111), and Nb-10 w/o W-10 w/o Ta, 3.96-mm (0.156-in.) - I.D. x 0.046-mm (0.016-in.) wall tubing, and various sizes of rod stock have been delivered for irradiation testing during the reporting period. The material was stress relieved for 2 hr at 850°C. The structural integrity of the tubing was determined by X-ray and eddy-current techniques.

b. Alloy Development. Arc-melt button castings have been prepared in the following compositions (in w/o):

Nb-15 Ti-6 V	Nb-10 Ti
V-5 Mo	Nb-10 V
V-50 Mo	V-40 Ti

Preparations for breakdown fabrication of the above alloys are now under way.

c. Duplex Tubing. Two 5.08-cm (2-in.)-diameter Type 304 stainless steel/refractory metal billets have been mandrel extruded at 1200°C on the 350-ton vertical press. One billet contained uncompacted high-purity niobium granules; the other contained a drilled bar of EB melted vanadium. Evaluation has not been completed, but initial metallography has indicated full compaction of the niobium and uniform tube blanks of both extruded products. The tube blanks will be conditioned and examined further by non-destructive means. Good sections will be processed further into small-diameter, thin-wall tubing.

2. High-temperature Metal Fuels

a. U-Mo Alloys.

(i) Casting. Further attempts to vacuum cast a U-14 w/o Mo alloy in a multicavity mold have been only partially successful. Six full-length castings, 1.27 cm (0.5 in.) in diameter and 30.5 cm (12 in.) long, were made, but some areas of porosity were revealed by X-ray radiographs

of the cast rods. Proper venting, coupled with other minor modifications in casting techniques, may be necessary to produce satisfactory castings of this alloy material.

(ii) Extrusion and Rolling. Unjacketed (bare) billets of U-14 w/o Mo have been extruded at a 4:1 ratio from a Houghton 1550 salt bath at temperatures of 750, 870, and 1000°C. Longitudinal striations and superficial circumferential cracks were noticed on the surfaces of the materials extruded at 750 and 870°C. The surface of the 1000°C extruded product was free from striations but more heavily cracked. Bare extrusion appears feasible, but proper lubricants must be found if the as-extruded surface is to be improved.

Three bare billets of U-14 w/o Mo were rolled to a total reduction of 67% from a Houghton 1550 salt bath at temperatures of 950, 970, and 1030°C with little real success. Surface cracking became a problem, being more extensive at the higher temperatures. Rolling to this reduction or higher can be satisfactorily accomplished with clad or jacketed billets between about 1000 and 1100°C.

b. U-Pu-Fz Alloy Fuel Rods. Thirty U-15 w/o Pu-10 w/o Fz EBR-II-type fuel pins containing 7.7 w/o U^{235} have been cast. These were radiographed by means of a Co^{60} source. Three of these have been machined to length and their densities determined.

Seven of the above specimens have been loaded into sodium-filled niobium and vanadium fuel tubes. Bonding will be performed upon completion of impact bonding equipment. Eddy-current testing for bond integrity requires a control cabinet. One such cabinet is now about 70 percent complete. The glovebox atmosphere with about 0.01 percent O_2 was found to damage niobium jackets when heated to 500°C for 4 hr for bonding. Results of tests made in a welding-grade atmosphere are being evaluated.

c. Development of Nonpyrophoric Uranium-Plutonium Fuel. A fuel alloy having a U^{238} -to- Pu^{239} atom ratio in the neighborhood of 2:1 would be a desirable fuel for a plutonium-fueled fast critical facility. This alloy, however, is very pyrophoric and also difficult to fabricate.

The French plan to use a U-25 w/o Pu-1.5 w/o Fe fuel in their Mazurka facility (Large Plutonium Critical Facility) and have reported some of their studies of U-Pu-Fe alloys with 0.5 to 1.5 w/o Fe.² They reported that the addition of 1.5 w/o Fe (≈ 6 a/o Fe) improves machinability and reduces pyrophoricity.

²French Progress Report - Secper-SMPu 592/63/3176 - 6/27/1963.

We are studying the effect of several alloying elements upon the machinability and pyrophoricity of the desired U-Pu alloy. Specimens are also being prepared for ignition temperature studies and possibly for isothermal oxidation studies. These specimens are being cast and machined into right cylinders, 6 mm in diameter and 6 mm long (0.240 in. in diameter by 0.240 in.) with a thermocouple hole drilled in one end. Duplicate specimens from the same castings will be exposed in an air-atmosphere plutonium glovebox in order to evaluate pyrophoric tendencies.

Cast specimens of U-27 a/o Pu with additions of 6 a/o Al, Cu, Fe, and Zr have been made, as well as control specimens of uranium and U-30 a/o Pu. The U-30 a/o Pu was difficult to machine as it fractured and crumbled readily. When exposed to the nitrogen atmosphere of the glovebox over the week end, specimens of this alloy powdered and crumbled (see Figure 3), even though the oxygen and moisture levels in the glovebox were kept below 300 ppm.

Figure 3

U-30 a/o Pu specimens, showing spontaneous disintegration that took place during one week-end in a nitrogen atmosphere with less than 300 ppm oxygen and 300 ppm water vapor.

With the addition of alloying elements, the machinability was greatly improved. The ternary alloys, however, were all very hard and tended to be brittle. They required a very sharp carbide tool for machining. The U-Pu-Fe machined well, giving a smooth surface. The U-Pu-Cu machined much like the U-Pu-Fe, but tended to chip more. It also had a porous surface when machined. The U-Pu-Zr gave a very smooth surface when turned, but fractured when drilled. The U-Pu-Al also gave a smooth surface but tended to break more readily from the tool pressure when turned rather than when drilled.

3. Nondestructive Tests of Tubing

a. Tungsten Tubing. Attempts to inspect seven pieces of tungsten tubing (nominally of 3.18-cm O.D. and 0.318-cm wall) for longitudinal defects with a conventional ultrasonic technique were unsuccessful.

No noticeable signals could be obtained from the reference V-notches on the I.D.; the larger of the two notches was 0.015 cm deep and 1.27 cm long. It is believed that the rough surface finish and coarse grain structure of the tubes severely attenuated the ultrasonic energy. Even at a frequency of 1 Mc the signals were weak and unintelligible.

However, it was possible to test these tubes for transverse defects at a frequency of 1 Mc. An I.D. V-notch, 0.01 cm deep and 1.27 cm long, again served as the standard. No indications equal to or larger than the standard indication were obtained. Several smaller indications appeared to be produced by machining marks on the surface.

b. Molybdenum Jacketed with Type 304 Stainless Steel. A piece of molybdenum jacketed with Type 304 stainless steel was inspected for bond quality by an ultrasonic through-transmission system at a frequency of 2-1/4 Mc. The transmit transducer was collimated to a diameter of 0.32 cm. A 0.16-cm-dia x 1.27-cm-long drill hole in one end of the rod served as the reference standard. Fairly uniform mechanical bonding was present in this sample.

c. Vanadium Rods. Two vanadium rods (nominally of 1.07-cm and 7.9-mm O.D.) were ultrasonically examined for transverse defects with a pulse-echo shear wave technique at a frequency of 1 Mc. Transverse V-notches, 0.01 cm deep and 1.27 cm long, were utilized as reference standards. One area in the 1.07-cm-O.D. rod produced a defect signal equal to the standard signal.

d. Vanadium-10 w/o Titanium Rod. Ultrasonic inspection of a vanadium-10 w/o titanium rod (nominally 3.40 cm in O.D.) for transverse defects was similar to that performed on the vanadium rods. The standard notch in this case was 0.08 cm deep and 1.27 cm long. No defects were noted.

e. Electromagnetic Tests. Electromagnetic tests of tubing being used or produced for general fast reactor fuel development gave results that are summarized in Table IV.

Table IV. Electromagnetic Tests of Tubing for Fast Reactor Development

Material	Dimensions		Quantity (m)	Results
	I.D. (mm)	Wall (mm)		
Hastelloy-X Lot No. 1	4.22	0.51	29.26 (2.44-m lengths)	One defect over 20% detected
Hastelloy-Y Lot No. 2	4.22	0.38	29.26 (2.44-m lengths)	Five defects over 20% detected
Hastelloy-X Lot No. 3	4.22	0.38	29.26 (2.44-m lengths)	No defects detected
Hastelloy-X	4.22	0.25	29.26 (2.44-m lengths)	Two defects greater than 10%
Type 304 Stainless Steel	4.22	0.18	5.2	No defects detected
	9.53	0.51	8.13	Numerous defects under 10%; 2 defects approx. 30%

These tests were made with pulsed-field test equipment and with the use of mask-aperture assemblies appropriate to the tube size.

At present it is necessary to scan tubes of greater than 6.6-mm O.D. spirally in order to obtain the sensitivity necessary to inspect tubes of 0.38-mm wall and thicker to a 10% defect level. Since this method is inherently very slow, experimental equipment is being designed which may possibly provide for high-speed inspection without loss of resolution and sensitivity.

f. Radiography of Hastelloy Tubing. Four lots of Hastelloy tubing of 4.22-mm I.D. were submitted for radiography. Each lot contained 12 tubes of 2.44-m lengths. Results are given in Table V.

Table V. Radiography of Hastelloy Tubing

Lot	Wall Thickness (mm)	Results
1	0.51	Tubes radiographically acceptable.
2	0.38	Numerous voids noted greater than 10% of the tube wall. These voids appeared in 9 tubes.
3	0.38	Voids greater than 10% of the tube wall noted in one tube.
4	0.25	Voids greater than 10% of the tube wall were noted in two tubes, remaining tubes radiographically acceptable.

All voids in the tubing are of the same type, appearing on the inner diameter of the tube wall. The size of the voids range from 0.51 mm to approximately 15.87 mm in length.

C. EBR-I, Mark IV

EBR-I continued to operate at design temperature and power throughout the month.

Additional Mark-III fuel elements were sent to Phillips Petroleum Co. for disassembly and storage for the impending reprocessing, bringing the total transferred to 112.

Additional tests were made on the charged-wire, cladding-failure monitor developed in conjunction with the EBR-I operation. The monitor uses the selective electrostatic fixation of daughter nuclides as a method

of discrimination against the A-41 normally present. The principle of operation is based on the fact that at the instant of beta decay each of the daughter species present becomes a positively charged ion. Accordingly, the cover gas is passed through an electrostatic precipitation unit which consists of a cylindrical shell and an axial wire. The cylinder is maintained at plus 2500 v. A closed-loop wire, driven axially through the cylinder, is maintained at ground. Each of the ionized daughter species resulting from beta decay migrates to the wire and becomes electrically neutralized. The wire carrying the neutralized daughters passes through a mercury trap-type gas seal which has a few cc of water floating on its surface. The strongly electropositive rubidium and cesium daughters react instantly with the water and are permanently fixed in the water layer in the hydroxide form. K-41 (the daughter of A-41) also reacts with the water; but, since K-41 is nonradioactive, the only activity associated with the water fraction is that arising from rubidium and cesium. Thus, a high degree of discrimination against A-41 is afforded.

The sensitivity of this device was tested rather conclusively during the past month. Enriched uranium foils, having a combined surface area equivalent to that of all four plutonium fuel slugs in a Mark-IV fuel rod, were irradiated at 900 kw. To facilitate the release of gaseous fission products, the foils were packed in a perforated basket. The basket was, likewise, installed in a perforated thimble located near the edge of the core, and the reactor brought to 900 kw in a conventional manner. The results of periodic count-rate measurements, corrected for the difference between Pu^{239} and U^{235} fission cross-section values, established a saturation value of 380 counts/sec approximately 5 hr after reaching 900 kw.

In contrast, previous measurements conducted under identical conditions of operation, but with no intentional exposure of fissionable material, resulted in a saturation value of 63 counts/sec. Subtraction of this value, a true measure of background effects, from the gross value of 380 counts/sec, results in a value of 317 counts/sec for the true failure signal. The signal-to-noise ratio, then, for the failure of a single Mark-IV fuel rod located at the edge of the core is $317/63$ or, approximately, 5.

From these results, it may safely be concluded that the present version of the charged-wire monitor is easily capable of detecting the failure of a single fuel rod in EBR-I. The results are particularly encouraging in view of the fact that detection of a failure is still possible in the presence of significant background fission products released continuously to the coolant from the vented blanket thermocouple rods in which uranium is exposed to the NaK coolant. The results suggest that the performance of a similar device would be even more satisfactory for a nominally clean loading.

D. EBR-II

1. Reactor Plant

During August the Reactor Plant was devoted primarily to accomplishing a procedural plan identified as R-14 for changing subassemblies from the loading used during sodium circulation and filtering to the pre-critical (non-nuclear) loading required for the start of the wet critical experiments. This reactor loading involved the following fuel-handling operations:

(1) Twenty-four subassemblies were repositioned in the core in order to place the natural uranium subassemblies in the fourth and fifth rows and move the dummy (steel) subassemblies into the third and fourth rows. The filters in rows one and two were left unchanged.

(2) In the inner blanket region, all 66 positions which contained filter and dummy subassemblies were emptied and then replaced with 64 standard inner blanket subassemblies and two antimony-beryllium sources.

(3) Three source storage thimbles were inserted in the outer blanket and three outer blanket subassemblies were removed.

The large amount of fuel handling involved enabled operating personnel to gain considerable experience and provided a good shakedown run for the fuel-handling equipment. Some of the typical difficulties encountered and corrected are the following:

(1) The fuel gripper, which was found to stick after continued operation, was freed by raising and lowering the gripper while manually rotating it. The apparent cause of the sticking is sodium oxide on the gripper shaft.

(2) Difficulties were encountered with the fuel-unloading machine gripper operation. This was partially due to oxide problems, as noted for the fuel gripper; also, the gripper caught on the leading edge of the bottom opening of the fuel-unloading machine. The oxide was removed by cleaning the gripper, and the bottom opening was chamfered to eliminate the latter fault.

(3) Galling had occurred in the storage-basket shaft, because of small clearances between the shaft and the packing gland spacer ring and follower. New parts were made with increased clearance to correct this condition.

(4) Miscellaneous electrical difficulties such as faulty switches and shorted wiring, were encountered. These were corrected as they were found. A recheck of the leak rate of all reactor building air locks and

freight door was in progress at month's end in preparation for wet critical. All building penetrations that had been worked on since the last leak test were leak-rate tested again.

2. Sodium Boiler Plant

The initial transfer of sodium from the storage tank to the secondary sodium-system piping, including the steam generator, was achieved on August 29 in 2 hr without incident. Following this, the sodium was circulated for nearly 1.5 hr and drained into the storage tank in 5 min. Plugging-temperature runs made after draining showed negligible change in the oxide content of the sodium. The storage tank was then heated from 350°F to 385°F. Plugging-indicator runs showed no effect on the oxide content.

The second sodium fill was started by pumping through the cold trap to reduce the sodium temperature to 350°F, the temperature of the system. Filling time was thereby increased to $3 \frac{1}{2}$ hr. After this fill, the main secondary EM pump was operated up to 50 percent flow or approximately 3,000 gpm. After draining the system, plugging runs again showed no significant oxide pickup.

Following the third sodium fill, attempts to increase the sodium flow resulted in tripping of the pump circuit breakers as a result of thermal overload at successively lower flow rates. After 23 hr, the system was drained; again the plugging indicator showed very little oxide pickup. The indicated plugging temperatures during this period of filling and draining were between 325 and 275°F. While waiting for the replacement thermal-overload devices, the sodium temperature in the storage tank was raised to 650°F. After reaching this temperature, the plugging temperature rose to 470°F. Overnight cold-trap operation reduced the plugging temperature to 350°F, which appeared to be an unreasonably quick response to cold trapping and indicated that sodium was probably channeling in the storage tank. In order to achieve better mixing, a temporary 2-in. pipe was installed from a flange near the cold trap outlet to a nozzle located on the far end of the storage tank. This line requires the sodium to travel the full length of the tank before returning to the purification system.

Prior to filling the secondary system, samples of the blanket gas in the storage tank were analyzed and found to contain approximately 150 ppm hydrogen. Following the third draining of the system, and before raising the bulk sodium temperature above 350°F, analysis of gas samples indicated the presence of 15 ppm hydrogen. This reduction in hydrogen is attributed to the formation of sodium hydride. After the temperature of the sodium-storage tank was raised to 650°F, the hydrogen content of the blanket gas increased to 280 ppm. This was attributed to the increased partial pressure of hydrogen from sodium hydride at the elevated temperature.

Sodium samples taken during this period showed small particulate matter in suspension. This has been identified as welding slag and mill scale, and is associated with the original condition of the secondary system piping when it was accepted from the construction contractor. During this period, the analysis indicated that the calcium content of the sodium had dropped to zero.

Following the repairs to the secondary EM pump circuit breaker, sodium in the storage tank was satisfactorily cold-trapped to 10 ppm oxygen. At month's end, a system heatup was in progress. An experiment was conducted to determine whether special action would be required to prevent steam generator damage in the event unit shell heaters are lost during the system heatup prior to filling.

3. Power Plant

The Power Plant piping had been altered to use feedwater heaters #3 and #4 during heatup for the initial filling of the secondary sodium piping system. By eliminating feedwater heater #2 and using closed-loop recirculation, demineralized feedwater makeup was reduced to a minimum. Upon completion of this phase of activities in the Sodium Boiler Plant, the Power Plant was placed on standby. Maintenance modification was resumed, and the valve for bypassing blowdown around the polishing demineralizer was replaced and repositioned.

The installation of exhaust relief valves on the turbines of the condensate and feedwater pumps is underway, and both relief valves are in place. The exhaust piping to the outside of the building still remains to be installed.

An intermittent leak in the bottom of the condensate storage tank was isolated and repaired.

At the end of September, feedwater heater#2 was placed in operation to heat the steam generators to 350°F for another sodium fill of the secondary system piping. The main steam line and the blowdown system have been prepared for full pressure operation at 580°F. The ultimate temperature and pressure will depend upon the amount of resistance and induction heating available in the secondary system.

4. Fuel Cycle Facility

Maintenance of quality and of pressure conditions for the Argon Cell atmosphere continued satisfactorily during the month. At the end of the period, the cell gas impurities were: oxygen - 45 ppm; water - 2.5 ppm; and nitrogen - 4.7%.

During overnight periods when the gas purification system is not in operation, the rate of air-inleakage into the Argon Cell has been periodically determined. With an Argon Cell pressure of two inches of water below atmospheric, the air-inleakage rate is now 0.005 scfm which may be compared with the previously reported (see Progress Report for August 1963, ANL-6780, p. 16) inleakage rate of 0.012 scfm.

A motor for one of the two atmosphere-circulating fans in the cell burned out, and was rewound and replaced. The fan vanes were reset to reduce the power required. The hoist and trolley drives of one of the two 5-ton cranes in the Argon Cell became inoperable due to an electrical failure within the cell. Preparations are being made to remove the trolley for examination and repair.

Repair of three Argon Cell manipulator bridge drives was effected by entry of personnel to the cell. The operation was carried out by teams of two men working on the extended platform of a forklift truck mounted on the hydraulic elevator platform in the transfer cell. The men wore Scott Air Paks with masks containing out-of-cell communications equipment. The entry operations resulted in the addition of about 4 percent of air to the cell. As expected, broken roll pins were found in four manipulator drives at the point of attachment of the clutch hub to the vertical drive shaft. These were replaced with solid pins. On one of the drives, complete repair was not possible at the time of entry because the pin could not be inserted fully. This will be completed later. Roll pins were also replaced on the two manipulators which were still in operating condition.

Minor modifications of the Argonne-fabricated interbuilding fuel-transfer coffin were completed. An operational shakedown test of the procedure for the removal of sodium from a fuel subassembly (see Progress Report for December 1962, ANL-6672, p. 20) was made with a clean (no coating of sodium), unirradiated fuel subassembly in the transfer coffin. The test results were considered satisfactory.

The spiral decanner and chopper is being used to prepare test charges for processing in the melt refining furnaces. This machine, which mechanically declads the fuel pins and chops them to a convenient length (about 2 in.) for handling, has operated satisfactorily. Fuel pins of depleted uranium-fissium alloy, with the alloy pins sodium-bonded to the stainless steel cans, have been provided for this purpose. After melt refining, the resulting alloy will be processed through the existing fuel fabrication equipment in the Argon Cell, and will be refabricated into sodium-bonded, canned fuel pins.

Initial in-cell operation of the decanning machine was undertaken, and removal of cans from about 50 elements was accomplished. Difficulties experienced were largely attributable to accumulation of sodium or its oxide on various surfaces. Sodium accumulated on the scrap compressor-cutter

gears, causing this unit to jam. A heavy accumulation of sodium also appeared to interfere with cutting-tool operation and with passage of cut fuel into the charger chute. A master-slave manipulator, being installed for servicing the decanner, should alleviate effects of these difficulties. Heating of components is also being investigated as a remedy.

Tubes for the temporary storage of spent and refabricated subassemblies are being installed in the Air Cell (see Progress Report for August 1963, ANL-6780, p. 17). Installation of a blower system for cooling the storage tubes and their contained subassemblies is being planned.

Two portable sample can cappers were received from the vendor and tested operationally. Following these tests, an inspection of the cappers revealed a need for corrective reworking of some of the components in order to eliminate the tendency of these parts to become scored.

A sample can and cover (both made of zinc) have been received from the vendor. The covered zinc can will serve as a storage container for oxidized skull material. Subsequently, the container and its contents will be charged to the skull oxide processing furnace. The wiring of the control system for the skull oxidation furnace (see Progress Report for February 1963, ANL-6698, p. 20) is being modified.

Distillation equipment for the removal of residual magnesium-zinc from uranium in the last step of the skull reclamation process (see Progress Report for July 1963, ANL-6764, p. 21) is being developed. Current work has been concerned with the development of a satisfactory joint between the condenser and collector to prevent sticking between these two components and to allow reuse of the condenser. The joint was modified to permit use of a graphite felt gasket. In runs in which 2 kg of magnesium-zinc were distilled, no sticking between the two components was encountered. However, the graphite felt gasket was found to bond to the condenser. Further modifications of the joint are necessary to make the equipment suitable for remote operation. Otherwise, difficulty would be encountered in the next run in seating the reused condenser (with attached graphite felt) on a new collector.

Several copper melts and two uranium melts have been made in the injection casting furnaces. In the first operation with uranium, the cast elements were short because of a temporary inadequacy of argon gas pressure. Elements of varying lengths were secured from the second melt.

It was found necessary to rewire the pin-processing machine because of interference between circuits. Rewiring of one machine has been completed, with the expected benefit. The fuel can welders were modified, and one is being installed in the Argon Cell. Assembly of leak-detector heads and of the bonder and bond tester was continued in the Air Cell.

5. Process Development

a. Remote-controlled Methods and Equipment for Fuel Refabrication. All equipment developed for remote refabrication of EBR-II fuel has been shipped to the site from ANL with the following exceptions: (1) the fuel-element assembly machine, (2) fuel-element tensile-testing and gaging machine, and (3) remote-controlled transfer grapples. The first two items remain in the machine builders' shops, and at current schedules will be ready for ANL inspection and testing in November. Grapples were requisitioned in August. Bids have not yet been received for their manufacture.

b. Skull Reclamation Process. The skull reclamation process will be used for the recovery and purification of the fissionable material that remains in the melt-refining crucible after a melt-refining operation. The steps include: noble metal extraction, uranium reduction, precipitation of a uranium-zinc intermetallic compound, intermetallic decomposition, uranium dissolution, and (after removal from the primary vessel to a separate vessel) retorting of the uranium product solution to concentrate the uranium. The steps are each followed by a phase separation in which a metal or flux phase containing impurities is separated from a uranium-bearing phase.

The phase separations are carried out by pressure siphoning. In a recent demonstration run with engineering-scale equipment, the efficiencies of the phase separations were further studied. A somewhat low removal of the supernatant liquid metal phase was obtained after the intermetallic compound precipitation. Since low removals of the waste supernatant from the intermetallic compound-precipitation step have been experienced previously, special efforts will be made to solve this difficulty. The efficiencies of phase separation obtained in the removal of the wastes after the other three process steps were satisfactory (> 90 percent).

All but the last of the process steps of the skull reclamation process are being conducted in a tungsten crucible. Because a flux heel remains in the crucible after uranium reduction, there is some residual salt in the crucible just prior to transfer of the uranium product solution to a separate vessel. This salt is in a molten state at the temperature of transfer and may be carried over with the product solution. It has been found in a small-scale run that by adding all of the magnesium fluoride required in the salt charge for the next run, the salt is maintained in a solid state and its transfer with the product solution is prevented.

Process operations in the engineering-scale equipment (see Progress Report for July 1963, ANL-6764, p. 21) have included four additional demonstrations of the skull reclamation process. Results have been obtained on the efficacy of the reduction of uranium oxides suspended in a molten halide salt to metal by a magnesium-zinc solution. Data indicate

that with effective mixing in this step, reduction of more than 99 percent of the uranium to metal can be realized in 2 to 4 hr at 800°C.

c. Sodium Coolant Chemistry. Experiments to evaluate a method for the determination of 10 to 100 ppm oxygen in sodium metal by inducing the $O^{16}(n, p)N^{16}$ reaction by means of fast neutrons were continued (see Progress Report for August 1963, ANL-6780, p. 24). The fast-neutron flux generated by means of a Cockroft-Walton generator was measured by irradiating a pure sodium sample for one hour and then counting the sodium-22 induced by a $(n, 2n)$ reaction. An effective flux of about $2 \times 10^8 n/cm^2\text{-sec}$ was measured. This value of the flux was used to estimate the nitrogen-16 activity which would be induced by irradiating sodium samples containing oxygen. The calculated activity values agreed within the experimental error with activity values experimentally determined by gamma counting with two 4-in. x 4-in. NaI(Tl) crystals whose estimated efficiency was only about 5 percent. A beta-detection system utilizing a small anthracene crystal was tested; results indicated that significantly higher detection efficiencies could be obtained with this system.

6. Irradiation of Prototype EBR-II Core-II Fuel Elements

An instrumented capsule containing six 7.62-cm (3-in.)-long prototype EBR-II Core-II fuel elements was recently removed from the CP-5 reactor after the fuel specimens had achieved a burnup of 2.4 a/o at 560°C. The fuel in each element is U-20 w/o Pu-10 w/o F's alloy clad with either Nb-4 w/o V alloy or Hastelloy X with a tungsten barrier layer.

Disassembly of the capsule revealed all claddings to be intact. No significant changes in diameter and density were observed. From neutron radiographs it was possible to measure the fuel elongation, which averaged 10 percent.

These specimens will be retained intact and will be either subjected to postirradiation annealing tests or re-encapsulated for additional irradiation.

7. Training

Activities for September involved 56 hr of training in preparation of reactor foremen for wet critical operation. This training covered the reactor auxiliary systems, reactor physics administrative procedures, and similar topics related to wet critical.

Nine new EBR-II technicians spent approximately two weeks at EBR-I, ZPR-III, and BORAX-V for training in the operation of these facilities.

Seven new technicians completed reactor technology classes, with the remaining new technicians having about two weeks of work remaining.

Four new EBR-II technicians completed plant orientation, with eight technicians having about one week remaining before completion.

E. FARET

1. Design Studies

The principal effort during September has been directed toward completion of the Title I design and the preparation of the Title I report. Meetings between Bechtel and Laboratory personnel have continued on an almost weekly basis. The design criteria for Title I have been completed, preliminary specifications for major equipment for procuring cost estimates have been completed and sent out to equipment vendors, and Title I drawings are nearing completion.

Bechtel has started a critical path analysis of the design and construction schedule for FARET, independently of that at the Laboratory. The initial Pert network from the Laboratory's study has been transmitted to Bechtel. The Laboratory's network is in the process of continued study, modification, and revision. The Title I design and construction schedule will be based upon Bechtel's schedule for the facility and the Laboratory's estimate for those portions of the plant being designed by the Laboratory.

The Title I report, due about mid-October 1963, will be a single volume containing a complete description of both the Bechtel and Laboratory portions of the design of the facility.

Stainless steels Type 304 or Type 316 have been chosen as suitable materials for the high-temperature sodium systems. For the purposes of the Title I design, it has been decided to use Type 304 for all locations where temperatures are expected to be below 1100°F and to use Type 316 elsewhere.

It was noted in the August Monthly Report (see ANL-6780, p. 26) that an evaluation of the containment requirements indicated that an increase in the pressure rating of the cell, cavity, and vault might be required. Further evaluation indicates that the pressure rating of the cavity and vault should be increased to the same value as that for the cell, i.e., 30 psig. On this basis, the Title I design criteria for the cell, cavity and vault are:

- a) The cell, cavity, and vault will be designed for a maximum pressure of 30 psig.
- b) Structural design stresses will be established at 80% of normal code values.

- c) Leakage from the cell, cavity, and vault enclosure will be no more than 0.1% of the total volume per day at a differential pressure of 1 psig. (This criterion is with all windows and other penetration closures in place.)

2. Safety Analysis

Work was begun by Electronic Associates, Inc. (EAI) at Palo Alto, California, on analogue studies of the FARET heat-transport systems for safety analyses. The work is under the direction of Bechtel Corporation. The Laboratory is participating to the extent of supplying all information required to program the reactor portion of the system.

At present, EAI and Bechtel have optimized the intermediate and secondary heat exchangers and piping for minimum numbers of zones. The reactor portion is programmed and the entire system representation should be operational by October 1. At this time, the following tests will be performed:

- a) scram of pump power from 50-Mw reactor operation, using main heat transfer loop only for natural-convection cooling;
- b) same as a) except only with the auxiliary heat transfer loop operating in natural convection.

For each of the above cases, eight runs are required: stepping of fission power to zero at four different times after scram, and using a high and low heat-exchanger elevation. Because of equipment limitations, both the main and auxiliary heat transport loop cannot be run simultaneously. Neither can the reactor kinetics or reactivity insertion equations be programmed. EAI has a large facility at Los Angeles which will accommodate the complete problem. After completion of early runs at EAI's Palo Alto facility, it is planned to perform the complete problem at Los Angeles.

An IBM-704 code (1922/RE) was written to calculate pressure vs. time in an enclosure following a sodium-air incident. The code was operated successfully in this reporting period. The code first calculates the peak temperature (and hence pressure) in the enclosure assuming:

- a) instantaneous reaction;
- b) stoichiometric combination of sodium with all the oxygen and water vapor in the enclosure;
- c) the same peak temperature for all the reactants.

The temperature profile is then calculated subject to the following assumptions:

- a) heat loss is to a liner by convection and/or radiation (gas radiation constants are not calculated);

- b) there are no heat losses from the gas-liner system;
- c) the liner and gas are at uniform temperatures at any time.

Various parameters are specified as input data. The output gives peak temperature and the profiles of gas temperature, liner temperature, and gas pressure (from the perfect gas law) as functions of time. The calculation ends when the liner and gas are at equilibrium temperature.

3. Oxygen Meter

Shop work is started on making the components for the oxygen-meter experiment (see Progress Report for August 1963, ANL-6780, p. 27). The thorium oxide electrolyte tube has been received, and most all other purchased items are now on hand.

4. Fuel Development

As noted in the Monthly Report for August (see ANL-6780, p. 29), tests and calculations indicate that the maximum change of fuel temperature in the test zone fuel with insulated-gas-gap fuel pellets of extruded and sintered UO_2 should be limited to 300-400°C if cracking of the pellets is to be avoided. Since a temperature change of at least 500°C is desired, efforts are currently being made to develop pellets which are less susceptible to cracking.

As a backup, fuel elements in which the pellets are a slip fit inside the clad are concurrently under consideration. With this type of element, which requires no development work, the fuel-temperature variation is obtained by relying on a large temperature difference through the fuel instead of across the fuel-clad gap. Since the thermal resistance of the gap is no longer important, and since the pellets are restrained by the clad, pellet cracking is not objectionable.

Changes of fuel temperature are accomplished by varying the reactor power together with the coolant flow rate to maintain coolant temperatures constant. Table VI indicates the calculated temperature changes obtained by varying the power and flow rate by a factor of 10 to 1. The fuel-pellet diameters of 0.5, 0.8, and 1.4 in. correspond to the use of 7, 3, and 1 fuel element(s), respectively, per subassembly. The values of fuel conductivity and gap conductance used are believed to bracket the ranges which can be expected.

Several conclusions can be reached by examination of Table VI and other related data. First, if the slip-fit elements are to produce larger changes of fuel temperature than permissible with the insulated-gas-gap elements, the diameter of the fuel pellets must be larger than 0.5 in. (The use of large pellets also reduces the importance of the fuel-clad interface temperature difference, which is probably more sensitive to fabrication tolerances and cracking variations than the fuel-temperature difference.) Secondly, it is possible to melt some of the fuel in 1.4-in.-diameter pellets, and perhaps even in 0.8-in.-diameter pellets.

Table VI. Change in Average Test Zone Fuel Temperature, °F(°C), due to Changes in Power and Flow Rate by Factor of 10 to 1

Slip-fit fuel; zoned core with 1058-liter driver; maximum reactor power of 50 Mwt; minimum reactor power of 5 Mwt.

Fuel thermal conductivity, B/hr-ft-F	1.0	2.0
Gap conductance, B/hr-ft ² -F	300	1200
Fuel element diameter, in.		
0.5	1105(615)	445(245)
0.8	2200(1220)	945(525)
1.4	5800(3220)	2630(1460)

Alternative materials based on UO₂ are metal-fiber reinforced UO₂ and UO₂ bonded with a silicate-base glass. Initial fabrication experiments with UO₂ powder plus 1 w/o sodium silicate produced pellets having a sintered density of about 94% of theoretical. These pellets had deformed to some extent in sintering and had developed a few small fissures. Additional work will be done to eliminate these defects, and to produce enough pellets for testing to see if this type fuel will withstand the heating rate without fragmentation.

Fabrication of UC pellets has started, for this material should withstand the heat fluxes anticipated in FARET better than UO₂. The UC was mixed with 1 w/o of stearic acid and milled in alcohol for 24 hr. Over 90% of the powder was found to have a particle size of less than 37 μ . This powder was compacted and sintered at 1650°C in vacuo. Compacts were found to have densities in excess of 90% of theoretical. Linear and volume shrinkages were determined and molds are being made to produce pellets suitable for testing.

5. Core Grid Structure

A structural analysis of the core supports for a conceptual configuration subjected to mechanical and thermal loadings has been completed. The analysis involves a thermal gradient which varies linearly through the thickness of the plate and is expressible by a convergent power series in which the radius r of the plate is the variable parameter. A general solution has been obtained for different rotational constraints. The solution has been applied to a sample model which is subjected to a combined static, internal pressure (see Figure 4) and thermal loading (see Figure 5). The temperature distributions shown in Figure 5 can be expressed as

$$T(r_1z) = B_0 + B_1r - \frac{z}{h} (A_0 + A_1r)$$

and the constants B_0 , --, A_0 , --, etc., for the top, middle, and bottom grid plates of the model are shown in Table VII where "a" denotes radius of the plate.

Figure 4. Mechanical Loads for FARET Core Support Model

Figure 5. Temperature Distributions for FARET Core Support Model

Table VII. Typical Loading Conditions for Sample Model

Plates	Static Load (lb)	Internal Pressure (psi)	Temperature Distribution Constants (°F)			
			B_0	B_1a	A_0	A_1a
Top Plate	90,000	10	0	37.5	0	-25
Middle Plate	0	75	25	30	-50	10
Bottom Plate	0	85	25	10	50	0

It is assumed that the grid structure is made of Type 316 stainless steel and is operated at 1000°F. The following material properties have been chosen for calculation:

$$E^* = 2.014 \times 10^6 \text{ psi}; \nu^* = 0.605; \alpha = 10.36 \times 10^{-6} \text{ in./in.-}^\circ\text{F},$$

where E^* and ν^* are the modified modulus of elasticity and Poisson's ratio, respectively, as defined for perforated plates by J. W. O'Donnell and B. F. Langer.

The diameter of each plate is taken as 66 in. The following pertinent quantities have been computed for each of the three perforated plates:

1. distribution of deflections;
2. maximum deflection vs. plate thickness;
3. distribution of radial stresses;
4. distribution of circumferential stresses;
5. stresses at center of plate vs. plate thickness;
6. radial stresses at edge of plate vs. plate thickness;
7. circumferential stresses at edge of plate vs. plate thickness.

The maximum deflections for three types of plate support are shown in Table VIII.

Table VIII. Maximum Deflections (in.) for Sample Model

Plates	Types of Plate Support		
	k = 0 (simply supported)	k = 0.5 (partially restrained)	k = 1.0 (clamped)
Top Plate (6 in.)	0.02	0.014	0.006
Middle Plate (4 in.)	-0.28	-0.18	-0.08
Bottom Plate (4 in.)	0.33	0.21	0.093

It is apparent that the actual support provided to the plates is somewhere between the two extreme conditions of simply supported and clamped. The partially restrained case gives a maximum deflection for the bottom plate of the order of $\frac{1}{4}$ in. To reduce the magnitude of the deflections and the thicknesses of the plates involved, other means for supporting the plates are being considered. Also, to improve coolant leakage aspects, the core support has been transferred to the bottom plate.

III. GENERAL REACTOR TECHNOLOGY

A. Theoretical and Applied Nuclear Physics

1. Elastic Neutron Scattering

A four-detector pulse-shape discrimination system, recently put into operation at the Van de Graaff facility of the Reactor Physics Division, was used to measure differential elastic neutron scattering cross sections at 8 angles between 20° and 160° . All angular distributions were fitted by the method of least squares to a Legendre polynomial expansion of the form

$$\frac{d\sigma(\text{el})}{d\Omega} = \sum_0^n b_L P_L(\cos \theta) \quad ; \quad \sigma(\text{el}) = 4\pi b_0$$

Results are reported for cobalt and chromium for incident neutron energies between 686 keV and 1097 keV. Measurements were made in

increments of ~ 20 -keV neutron energy, with a resolution of ~ 8 keV. All data have been corrected for effects due to multiple scattering.

The results for cobalt are given in Figure 6. The open circles are the experimental results averaged over 60-keV intervals. The solid curves are the results of an optical model calculation.

The results for chromium are given in Figure 7. Strong resonance contributions are evident in all of the coefficients. The resonance contributions in the b_4 coefficient indicate probable D-wave interaction.

2. Ultra-low-energy Proton-recoil Measurements

Some problems associated with the use of hydrogenous proportional counters for making proton-recoil measurements over the range of ionization from one to about 20,000 initial electrons have been studied. Evidence exists that

Figure 6. Elastic Neutron Scattering from Natural Cobalt. The neutron-energy resolution is ~ 60 keV. The open circles are the experimental points and the solid curves are the results of an optical model calculation.

Figure 7. Elastic Neutron Scattering from Natural Chromium. The neutron energy resolution is ~ 20 kev

it was observed that a plot of the form $(\log A)/V$ vs. $\log V^3$ was, within uncertainties, linear when A exceeded about 10. This is an important result since it permits an entire gain-voltage map to be drawn from a single set of data with one calibration source (which will undoubtedly be $N^{14}(n,p)C^{14}$ for proton-recoil work). No other reported test of this formula has covered the entire proportional region.

Single electrons ejected from the cathode by ultraviolet light multiply when within about one anode diameter (1 mil), and the resulting avalanche releases up to about 10^6 additional electrons, depending upon counter voltage. There are large fluctuations in the number of final electrons from event to event, however, and the pulse-height spectrum is broadly distributed. Theories of this distribution⁴ predict that it should be exponential,

pulse-shape discrimination will permit recoil protons to be separated from the accompanying background of gamma-induced events over most of this range subject to the restriction that the total gamma background rate is countable (all events which give rise to one or more electrons in the gas are detected).

In order to establish the variation of gas multiplication A with voltage and to fix the maximum value of A that can be tolerated without space-charge saturation, ultraviolet light was introduced into a counter containing methane gas with 5% nitrogen. Photoelectrons ejected from the cathode were counted individually to provide "single electron" calibration data. By exposing the detector to thermal neutrons, events from the $N^{14}(n,p)C^{14}$ reaction were also detected (corresponding to the release of about 20,000 initial electrons). Measurements of A versus voltage over the extremities of gas multiplication were made, and

³W. Diethorn, A Methane Proportional Counter System for Natural Radiocarbon Measurements, NYO-6628 (March 16, 1956).

⁴O. R. Frisch, Fluctuations in Chain Processes, AECL-748 (Feb 1959).

but the only published data⁵ is more consistent with $x^{1/2}e^{-x}$ than with e^{-x} . A single-electron distribution taken with $\text{CH}_4 + 5\% \text{N}_2$ at 1 atm is given in Figure 8 and over almost four decades results are consistent with a pure exponential.

Figure 8. Single-electron Pulse-height Spectrum

This is quite fortunate since it facilitates calibration with ultraviolet (the average pulse height is the reciprocal of the slope of the distribution). More widespread use of single electrons for calibration appears to offer substantial advantages for making measurements at the lower neutron energy levels.

3. High Conversion Critical Experiments

Measurement of reactor physics parameters continue in the core described in the Progress Report for July 1963, ANL-6764, p. 35. The

⁵S. C. Curran, S. L. Cockraft, and J. Angus, Investigation of Soft Radiation by Proportional Counters - Part V, Phil. Mag. 40 (1949).

U^{238} to U^{235} fission ratio δ^{28} has been measured; a value of 0.1160 ± 0.0016 was obtained from 16 combinations of depleted and enriched foils. Foils were irradiated at four radii in the stainless steel-clad zone. If $\nu^{25} = 2.43$ and $\nu^{28} = 2.84$, then a value of $\epsilon = 1.088 \pm 0.0012$ is inferred from the value of δ^{28} . Analysis of results of measurements for the determination of the internal conversion ratio and the U^{238} capture cadmium ratio is in progress.

For this lattice the measured values for the U^{235} fission cadmium ratios are:

in the fuel	3.90 ± 0.14 ;
in the moderator	3.93 ± 0.14 .

The epicadmium to subcadmium fission ratio for U^{235} , δ^{25} , is then

$$\delta^{25} = 0.345 \pm 0.017$$

The thermal activation technique developed by Babcock and Wilcox (see BAW-1268) was used to measure the U^{238} capture cadmium ratio, C_{28} , in this system, giving $C_{28} = 1.191 \pm 0.009$. The ratio of epicadmium to subcadmium captures in U^{238} is then $\rho^{28} = 5.23 \pm 0.25$. The primary advantage of this technique for measuring ρ^{28} is that $C_{28} - 1$ is measured directly instead of C_{28} ; thus, the percent error in ρ^{28} is reduced to simply the error in the measurement of $C_{28} - 1$.

A pneumatic sample oscillator was installed in the Hi-C facility to test the feasibility of measuring the capture to fission ratio of U^{235} by an oscillation technique. The results indicate that meaningful data can be obtained with a spontaneous fission source (Cf^{252}) strength of the order of 10^4 n/sec; however, an increase in source strength by a factor of 10^2 would be desirable.

The temperature coefficient of reactivity was measured for the 1.27-cm triangular, aluminum-clad Hi-C lattice. For the range from 20 to 40°C an average value of $-1.01 \times 10^{-4} \Delta k/^{\circ}C$ was observed. It can be noted that this is not substantially different from the value $-1.04 \times 10^{-4} \Delta k/^{\circ}C$ observed for the 1.24-cm square, stainless steel-clad Hi-C lattice (see Progress Report for November 1962, ANL-6658, p. 36).

4. Theoretical Physics

a. Matrix Representation of Detection Systems. Since ideal detection systems do not exist, the measurement of a spectrum of physical interest is often complicated by the necessity of determining the actual spectrum from the observed experimental data. This unfolding problem can be conveniently formulated in terms of a matrix representation. In

this approximation, one has

$$\underline{Y} + \underline{E} = A\underline{X} \quad , \quad (1)$$

where $\underline{Y}$, $\underline{X}$, and $\underline{E}$ are n -element column vectors and A is an $(n \times n)$ matrix. Here $\underline{Y}$ represents the output data of the detection system and $\underline{E}$ is the inherent experimental error in the measurement. The matrix A is commonly called the response matrix of the detection system.

The solution of Eq.(1) for the unknown vector $\underline{X}$, which represents the desired spectrum, can be written as

$$\underline{X} = A^{-1} (\underline{Y} + \underline{E}) \quad , \quad (2)$$

where A^{-1} is the inverse matrix of A . The crux of the present problem stems from the fact that the solutions determined by Eq.(2) are nonunique. This nonuniqueness property, which arises from the existence of the error vector $\underline{E}$, is complicated by the fact that most response matrices of interest are ill-conditioned. Thus, instead of a unique solution, one generally finds an infinite set of vectors

$$\chi = \{ \underline{X} \mid \underline{Y} + \underline{E} = A\underline{X} \} \quad (3)$$

which satisfy Eq.(2). Consequently, one must employ subsidiary conditions to determine which element of χ is the desired solution. For example, smoothness criteria have been utilized with some success.⁶ We are investigating an iterative method of solving Eq.(1) which inherently utilizes many physical implications as subsidiary conditions. The iterative method has been programmed, and the convergence properties of this method are under investigation with the aid of the IBM-704 computer.

Although the solutions determined by the iterative method have been adequate in some instances, the rate of convergence is poor. It appears that the rate of convergence depends on the choice of the initial vector for the iteration as well as the "conditioning" of the response matrix. Methods to increase the rate of convergence will be examined.

b. Importance-weighting Approximation Method for Reactor Analyses. An importance-weighting approximation procedure has been formulated to calculate few-group parameters for diffusion theory computations of k_{eff} in multiregion source-free reactors. The formulae define a mechanism of group expansion, reducing the number of neutron energy groups, while accounting for the variation of the original multi-group flux spectrum near regional boundaries. The resulting few-group equations could be applied to survey-type computations for k_{eff} , or to two-dimensional analyses of reactors of similar configuration and composition.

⁶D. L. Phillips, J. Assocn. Computing Machinery, 9, 84 (1962).

Let the basic description of reactor equilibrium be provided by a system of m diffusion equations of the form

$$0 = D_j \nabla^2 \phi_j - \Sigma_{R,j} \phi_j + \sum_{k=1}^m \phi_k \Sigma_{\text{source}}(k \rightarrow j) \quad ; \quad 1 \leq j \leq m \quad , \quad (4)$$

in each region of the reactor. Let the unnormalized solutions of Eq.(4) be ϕ_j . Denote the corresponding unnormalized solutions of the adjoint system by ϕ_j^* :

$$0 = D_j \nabla^2 \phi_j^* - \Sigma_{R,j} \phi_j^* + \sum_{k=1}^m \phi_k \Sigma_{\text{source}}(j \rightarrow k) \quad . \quad (5)$$

In each region of the reactor, define the total weighted few-group reaction rates:

$$\begin{aligned} \bar{\Sigma}_{R,p} \int_{\text{region}} \Phi_p \Phi_p^* d\underline{r} &\equiv \sum_{j \in G_p} \int_{\text{region}} \Sigma_{R,j} \phi_j \phi_j^* d\underline{r} \quad ; \\ \bar{\Sigma}_{\text{source}}(l \rightarrow p) \int_{\text{region}} \Phi_l \Phi_p^* d\underline{r} &\equiv \sum_{j \in G_p} \sum_{k \in G_l} \int_{\text{region}} \phi_k \phi_j^* \Sigma_{\text{source}}(k \rightarrow j) d\underline{r} \quad ; \\ \bar{D}_p \int_{\text{region}} (\nabla^2 \Phi_p) \Phi_p^* d\underline{r} &\equiv \sum_{j \in G_p} \int_{\text{region}} D_j (\nabla^2 \phi_j) \phi_j^* d\underline{r} \quad . \end{aligned} \quad (6)$$

In Eq. (6), $\Phi_p \equiv \sum_{j \in G_p} \phi_j$ and $\Phi_l^* \equiv \sum_{k \in G_l} \phi_k^*$, the summation running over all fine groups of the corresponding coarse group; $1 \leq l \leq n$, and $1 \leq p \leq n$.

Then, in the weighted mean,

$$0 = \bar{D}_p \nabla^2 \Phi_p - \bar{\Sigma}_{R,p} \Phi_p + a \sum_{k=1}^n \Phi_k \bar{\Sigma}_{\text{source}}(k \rightarrow p) \quad , \quad (7)$$

where "a" is the constant factor which is required for the satisfaction of Eq. (7).

When the m -group real fluxes are separable into a simple product of a function of space and a function of energy, Eq. (7) is solvable

with $a = 1$. This means that the few-group value of k_{eff} is the same as the multigroup value. For such an idealized case, the operation of group expansion is transitive, in the sense that successive applications of group expansion lead to the same equations as a single direct reduction in the number of groups. Automatically, in this case, the weighted few-group reaction rates integrated over the region are identical with the corresponding weighted multigroup reaction rates integrated over the region.

For the typical case in which the fluxes are not separable, the importance-weighting method only approximately preserves the multigroup value of k_{eff} . This procedure will be applied to the computation of k_{eff} for a number of reactors of different types to test its usefulness.

c. Development of Reactor Computing Codes

(i) Resonance Interference. The code 1559/RE has been rewritten in the FORTRAN 60 Language for use with the CDC-3600 Computer. The first part of the program writes a library tape which may then be used for a large number of problems based on the second part. Execution time for a library tape of 2 isotopes, 7 levels, and 56,000 lethargy points required 3 min on the CDC-3600 as compared with 30 min on the IBM 704.

(ii) SLOP-1. Libraries 1 and 2 have been written and are being processed for use with SLOP-1. These libraries have a 19-group structure and include all the isotopes on the SOFOCATE list. Library 1 is for 21°C, number 2 is for 100°C. The Radkowsky kernel is used.

d. Fast Reactor Control. Exploratory calculations have been initiated to specify parameters relating to control of large (>1000-liter core volume) fast power breeder reactor systems. As expected, it is difficult to obtain large reactivity swings with nonparasitic absorbing control mechanisms. The latter conclusion is most evident if the core volume donated to control mechanisms is to be minimized. A most interesting result pertains to natural boron substitution. It was found that a mixture of natural boron and hydrogen (at about water density) is about 30% more effective than the same volume occupied by natural boron. This reactivity may be enhanced by ~10-15% in some dilute systems if the hydrogen-bearing material can be caused to move separately from the boron.

The introduction of hydrogen causes some spectral degradation, at least in the vicinity of the control rods. Small substitutions giving ~1-2% $\Delta k/k$ do not significantly affect neutron economy.

e. S_N Calculations. Currently there are several versions of the S_N Method available for reactor analysis. These include SNG, DSN and DTK. In particular, the SNG version is used for the neutronic subroutine in the AX-I program. Some reactivity determinations as part of the

AX-I calculation were found to be in poor agreement with previous DSN analyses. To resolve the differences, a series of controlled reactor calculations were initiated to compare DSN with SNG, and hence with AX-I. It was found that SNG and DSN were in substantial agreement for a heavy metal core with a thick, heavy metal reflector. However, the use of a thick, moderating and nonabsorbing reflector appears to be the cause of the difficulty cited above. For the latter situation it is possible to obtain a difference of $\sim 10\% \Delta k/k$ between a DSN and SNG calculation.

B. Reactor Fuels Development

1. Corrosion Studies

a. Zirconium Alloys for Superheated Steam. Detailed studies of corroded samples of Zr-Ni-Fe and Zr-Cu-Fe alloys have been continued. The results of microhardness traverses on specimen cross sections support metallographic evidence that oxygen diffusion into the metal is appreciable in the steam environment at 650°C and 42.4 kg/cm^2 . If crazing of metal around the DPH indentation is used as an index of embrittlement, the material may be said to be brittle at DPH values higher than 550 (at 100-gm DPH load). Several hot-rolled and heat-treated samples, corroded for 88 days at 650°C , displayed this hardness value at depths ranging from 40 to 70μ below the film-metal interface. The DPH-versus-depth curves fell to within 10% of bulk metal hardness (200 DPH average) at depths of 250-290 μ (10-12 mils). For comparison, in similar samples corroded for 300 days at 540°C the hardest point observed was 480 DPH at 17 μ from the metal surface, and the curves fell to within 10% of bulk value at 48-92 μ (2-3.5 mils). All samples at both temperatures were much thicker than the affected depths, and the bulk hardness values observed were not significantly higher than before corrosion test. One sample of Zr-2 w/o Fe showed no gradient of hardness inwardly from the metal surface after 650°C exposure, but this was attributed to its exceptionally high corrosion (weight-gain) rate.

Results of several rather questionable DPH readings of the 650°C corrosion films yielded values averaging 860. The 650°C -hardness curves for the metal may readily be extrapolated to this range by extending the curves smoothly through zero metal depth (although considerable freedom exists due to point scatter in this region). Thus, the film-metal interface may be merely a phase boundary in the zirconium-oxygen system and not a sharp boundary between an outer, rate-limiting film and the inner metal; that is, the corrosion behavior of these alloys after the beginning of exposure to steam may be related to diffusion behavior in the metal more than through the film. Such behavior is in agreement with previous hypotheses. Whether or not this mechanism may be extended to steam corrosion at 540°C is being considered.

Other corrosion tests of binary alloys, as-cast materials, and heat-treated materials in 540°C and 650°C, and 42.4-kg/cm² (600-psi) steam are being continued. Apparatus is being prepared for similar tests at as low as 1 torr pressure of water vapor to (a) determine the effect of pressure, and (b) relate electrical surface phenomena at low pressures (previously described) to corrosion at higher pressures by perhaps demonstrating a lack of pressure effect for these zirconium alloys.

b. Ferrous Alloys in Superheated Steam. The dynamic test of Types 304 and 406 stainless steel (see Progress Report for July 1963, ANL-6764, p. 41) continued for 82 days' exposure and will continue for an additional three months. Fresh samples with a vapor-blast finish have been used to replace those removed for defilming in order to provide information about the long-term corrosion stability of the vapor-blasted surfaces.

A refreshed static test in (30 ppm O₂) steam at 650°C and 100 psi is being used to evaluate the effect of pressure on the corrosion rates of Type 304 and Type 406 steel.

Work continues on the new autoclave system for tests at higher temperatures.

2. Ceramic Fuels

a. Urania Burnable Poisons. The study of the solid solution formation in compositions of urania and gadolinia sintered at 1700°C for 4 hr in argon and in hydrogen continues (see Progress Report for August 1963, ANL-6780, p. 36). Sintering in hydrogen produced a face-centered cubic solid solution of urania and gadolinia up to at least 80 m/o Gd₂O₃. There is an anomaly in the behavior of the lattice parameter at 40 m/o Gd₂O₃. The lattice parameter decreases linearly from 5.4668 a.u. for UO₂-5 m/o Gd₂O₃ to 5.4010 a.u. for UO₂-40 m/o Gd₂O₃. Above 40 m/o Gd₂O₃, the lattice parameter decreases, at a slower rate, to 5.3827 a.u., for UO₂-80 m/o Gd₂O₃.

An analysis of the X-ray diffraction patterns for the hydrogen-sintered materials indicates the presence only of the face-centered cubic solid solution from UO₂-5 m/o Gd₂O₃ to UO₂-80 m/o Gd₂O₃. At this latter composition, extraneous lines, as yet unidentified, are found on the diffraction patterns.

b. (U, Th, Pu) - P Systems

(i) Preparation of Plutonium Phosphide. A number of batches of plutonium phosphide were prepared at different temperatures. The best product, as determined by X-ray diffraction, was obtained between 650 and 685°C. Samples of PuP are now being analyzed for phosphorus and nitrogen content.

(ii) Uranium Phosphide-Uranium Sulfide. Studies of the UP-US system are continuing. Sintered samples with a composition toward the US end of the system exhibited considerable cracking, while the US sample itself was sound. These cracks showed up again in samples of the same composition during a second run and are attributed to some outgassing reaction. On melting, the cracked samples appeared to exhibit higher vapor losses than the uncracked specimens. This phenomenon will be studied further

Measurements of the room-temperature electrical resistivity of samples showed an initial increase in resistivity when US was first incorporated in UP. Following this, the values decreased with increasing US concentrations. The cracking of sintered specimens toward the US end of the system does not permit accurate resistivity measurement of these samples. The electric resistivity of US was $260 \Omega \text{ ohm-cm}$ as compared with $320 \Omega \text{ ohm-cm}$ for UP. These values were corrected for the porosity factor.

Oxidation studies were conducted with powdered samples in the UP-US system by differential thermal analysis (DTA). US exhibited a sharp exothermic peak at about 380°C , whereas UP exhibited a broad peak at about 550°C . Addition of 20% UP to US shifted the exothermic peak to 500°C and made it considerably broader. Addition of 40% US shifted the peak to 550°C .

The incorporation of UP in these amounts is enough to retard oxidation by the formation of a vitreous P_2O_5 coating around the grains. The coating is not effective above 550°C since P_2O_5 melts at this temperature. Residues from the DTA runs of all samples, except that of US, contained the compound $(\text{UO}_2)_2\text{P}_2\text{O}_7$ in addition to U_4O_9 . All of the samples were heated to 1000°C in a flowing atmosphere of oxygen.

c. Plutonium Sulfide. An apparatus to be used for preparing the compounds of plutonium has been constructed and installed in a glovebox. The first method of preparation of plutonium sulfide is the same as was used to prepare US: uranium metal is hydrided several times to make a fine powder, which is then reacted with hydrogen sulfide to make US. Preliminary work on the hydriding and dehydriding of plutonium showed that it is difficult to prepare a plutonium metal powder from the hydride. Some large pieces were still left after dehydriding and the dull luster of the product indicated the presence of some hydride phase. Reaction of this material with H_2S was tried at temperatures from 300 to 580°C . The gas pressure was monitored as an indication of the rate and extent of the reaction. Below about 500°C the reaction was very slow. At 580°C it occurred readily; however, examination of the apparatus showed that a thin film of product had vapor-deposited on the walls of the reaction chamber. Work is now in progress to identify the various reaction products by X-ray diffraction. When these results are available they should provide the information that is needed in order to choose the best reaction temperatures.

d. Uranium Monocarbide. Specimens of UC and 10 w/o US-UC containing metal particles as a dispersed phase were heated in vacuum to obtain comparative compatibility information. The UC used was arc melted and ground to a particle size of $<15\mu$; it contained 4.8 w/o carbon and 0.4 w/o oxygen. The US was of similar particle size and contained 11.76 w/o sulfur and 0.1 w/o oxygen. The US-UC composition was pre-reacted in vacuum at 1600°C and reduced to $<15\mu$ particle size by grinding. Specimens were pressed from the powders mixed with about 20 v/o metal filling and 1% binder. The specimens were heated in vacuum at 1150°C for 192 hr. This temperature was necessary to obtain some sintering of the powders so the specimens could be mounted and polished, and also to insure reaction in a relatively short period of time. The severity of the reaction between the metals and the compounds was observed by means of metallographic examination, and the results are listed in Table IX.

Table IX. Compatibility of Refractory Metals with UC and 10 w/o US-UC

Metal	UC	10 w/o US-UC
Type 304 Stainless Steel	Complete reaction or migration of metal; densification of specimen much greater than others	Metal particles still intact, although some reaction as evidenced by carbide crystals grown in metal
Niobium	Complete reaction of smaller metal particles; larger particles contained metal cores	Very slight reaction at surface of metal particles
Tantalum	Complete reaction of particles with carbide matrix; appears to be two carbide phases in particles	Shallow reaction zone with core
Molybdenum	Complete carburization of metal particles	Very slight reaction if any
Vanadium	Complete carburization	Partial reaction; some metal left

Only in the specimen of UC containing Type 304 stainless steel did the metal particle lose its identity as a particle. In all of the other specimens the particles maintained their shape, and the reaction took place by migration from the matrix.

e. Uranium-Plutonium Carbide Fuel. A total of 16 vibratory-compacted carbide specimens were fabricated during the reporting period. Two problems were encountered: contamination of the welds, and defective

welds. Both defects were found to be related to procedures used for decontaminating the tubes after filling but prior to closure. The procedure was changed, and in the last ten specimens only one showed contamination greater than 300 dpm after welding.

The current procedure is as follows: A close-fitting teflon-on-nylon fitting is placed over the open end of the specimen tube. The fitting extends down into the tube and forms a funnel through which the powdered carbide is loaded. The outside of the tube is coated by dipping in a collodion-ether solution. The tube is placed in a secondary container which is sealed against the teflon (or nylon) funnel. Filling and vibration compacting is done within this secondary container to keep alpha contamination from the outside of the tube.

After filling and compacting, the protective container is removed and the tube is transferred to a low-contamination area. Here the filling fitting is carefully removed and the top plug inserted. The collodion is stripped and a preliminary contamination survey is made. If the tube is free of contamination, a weld is made and a final survey is made for loose and fixed activity.

It has been found necessary to keep both collodion and "wet" decontaminating media from contacting the weld zone prior to welding. Collodion was formerly used to seal the loading fitting to the specimen tube. With the current technique, the upper $\frac{3}{8}$ in. is not coated. Only dry cleaning methods are used. Fiberglass erasers effectively remove the collodion coating. Efforts are now being made to simplify and speed up the operation, which is still too tedious for large-scale production.

3. Particulate Metal Fuel Elements

The investigation of methods for producing powdered metal and nonmetal fuel elements is divided into two phases: (1) production of U-Pu-Fz metal powder and (2) development of techniques for compacting this powder with controlled density into specimen jackets.

a. Powder Production. Both atomizing and pulverizing techniques are being investigated for powder production. A total of 12 atomizing melts have been made to date. The maximum yield of finely divided powder has been 28 percent. The screen analysis of the material is shown in Table X, Column "b." The balance of the material cast forms a spongy metal deposit in the collector tray. The rotary collector, after casting, is shown in Figure 9. The finely divided particles collect at the outer rim of the centrifugal collector. Figure 10 shows an "icicle" of fuel alloy which developed on the nozzles of the atomizer crucible.

Table X. U-10 Pu-10 Fz Alloy Powder

(a)	(b)	(c)	(d)	(e)
NBS Screen Mesh	Atomized	Atomized and Pulverized		Cast and Pulverized 437-g Charge
		98-g Charge	999-g Charge	
on 14	47.2%	1.6%	-	-
14-45	17.8	28.0	4.2%	1.7%
45-120	5.1	47.0	56.6	34.3
120-200	1.5	13.0	25.0	28.2
200-325	1.6	6.4	8.6	23.8
minus 325	1.7	4.2	5.6	12.0

Figure 9. Rotary Collector after Casting

Figure 10. "Icicle" of Fuel Alloy on Nozzles of Atomizer Crucible

The spongy residue was then pulverized in a hammer mill (micropulverizer). Essentially all of it was broken up into fine powder, as shown by the screen analysis in Table X, Columns "c" and "d."

In view of the excellent results achieved with the pulverizer, some injection-cast pin ends were fed through the micropulverizer. These, too, were powdered with little difficulty, as shown by the screen analysis in Column "e" of Table X.

Figure 11 shows shadow photomicrographs of the powders produced by atomizing and by pulverizing.

b. Vibratory Compaction. Since the last reporting period a series of runs with binary mixtures of steel shot have been made. The ratios of large-diameter to smaller-diameter shot were held constant, and maxima in the packing fraction were found to exist. The nature of the relationship between the packing fraction and the ratio of diameter of tube to diameter of the larger-sized fraction has been determined. The

120 - 200 mesh

200 - 325 mesh

- 325 mesh

(a) by pulverizing

(b) by atomizing

Figure 11. Powders Produced (Magnification 100X)

results are somewhat erratic. We now have evidence that the spurious results are brought about by "bridging" of the smaller-diameter spheres within the interstices of the matrix formed by the larger-diameter spheres. Vibration in a plane normal to that of the driver-imparted vibration serves to dislodge the "bridged" particles and promotes compaction. Work is now in progress to determine maxima with the aid of an additional mode of vibration.

4. Extrusion Development

a. Production of Molybdenum Tube Blanks. Five extrusions of molybdenum tube blanks have been made during the past month. All billets contained molybdenum sleeves protected by Type 304 stainless steel as an outer jacket and contained cores of Type 304 stainless steel. Three billets incorporated an Al_2O_3 -filled annulus between the ID of the molybdenum sleeve and the core (modified DPM technique). Two of these modified DPM billets were extruded at 1200°C . The remaining billets were heated to 1100°C and delayed in transfer to the press until the temperature of the outer can surface was approximately 1000°C .

All billets extruded satisfactorily and X-ray radiography indicated uniform molybdenum tubing in the extrusions. The ID surfaces of the molybdenum sleeve appeared slightly roughened by the Al_2O_3 powder. This condition was expected and is not considered detrimental to secondary fabrication. In the billets not having the powder-filled annulus, the 304 stainless steel core shrank away from the molybdenum sleeve because of differential thermal expansion. Core removal after either method of extrusion is not difficult.

The molybdenum tube blanks produced by the two techniques will be dejacketed, nondestructively tested and further processed.

5. Nondestructive Testing

a. Neutron Imaging. The possibility of employing a phosphor screen to perform neutron fluoroscopic inspection has been investigated briefly. In the direct neutron beam, several phosphors yielded an image bright enough for visual observation of the beam shape. The best phosphor of those examined was the 4:1 by weight mixture of $\text{ZnS}(\text{Ag})$ and Li^6F , which had previously been found to yield the fastest photographic neutron response.

Attempts to use the fluoroscopic technique for the inspection of uranium and other heavy materials have been unsuccessful, however, because neutron-induced prompt-emission radiation in the inspection material also influenced the light emission from the phosphor. This may offer some explanation for the relatively poor sensitivity to radiographic contrast obtained for neutron inspection when phosphor-film detectors were used. Some further work on this aspect of the study is being done in order to clear up this point.

The neutron facility has been employed to inspect a number of pieces of 7.5-cm-diameter uranium-molybdenum alloy rods. The total length of material inspected was 360 cm.

Other work completed this month includes two inspections of radioactive material, one of these being an experimental reactor fuel capsule irradiated at CP-5 and the other an experimental reactor control assembly irradiated at MTR.

b. Ultrasonic Imaging. Camera-circuit modifications necessary for the operation of the sealed-off ultrasonic camera tubes have been essentially completed. Several tubes have now been operated with this equipment. Further modifications may be necessary to improve focusing arrangements when the tubes are operated with an ultrasonic signal. A water tank is now being set up to permit these studies to continue.

The continuously pumped ultrasonic camera system has been used for a resolution study with metal test objects with holes all the way through them. With thin metal plates (thicknesses less than the wavelength λ of the ultrasonic beam in water), the depth of focus was about 3 cm for direct shadowing methods; that is, holes which varied in diameter from λ to about 10λ could be well imaged when the object was moved from direct contact with the detector to a distance of about 3 cm. Beyond this distance the hole images became appreciably less sharp.

With the use of thick plates (thickness equal to 10λ), hole sizes equal to or greater than 5λ could be well imaged within a distance of about 5 cm from the detector. Hole diameters less than 5λ showed the influence of diffraction effects. Although these hole sizes could be well imaged at object-detector distances up to 7.5 cm, it was difficult to correlate image size because of diffraction effects.

Resolution studies are continuing with lens systems and metal test objects containing flat-bottomed holes, drilled part-way through.

c. Correlation of Heat-transfer Properties and Bond Quality. Both the physical and the electronic apparatus for the flash-tube method of measuring thermal diffusivity which is being utilized to determine heat-transfer properties of small bonded samples have been completed. At present, the system is being checked out to insure that the experimental temperature-rise curve agrees with the theoretical curve. The effect of an optical system and various methods of attaching thermocouples to the specimens are also under study.

d. Ultrasonic Instrument and Transducer Development. Lead metaniobate and lead zirconate titanate (Clevite's PZT-5) were studied as crystal materials for ultrasonic transducers. The lead metaniobate crystals

were obtained from Branson Instruments. One lead metaniobate transducer has excellent sensitivity plus a fairly short pulse length ($\sim 4\mu$ sec). The PZT-5 transducer appears to be better suited for passive or receiving applications.

Equipment for a Schlieren optical system has been ordered. This system will be used to analyze the radiation patterns of transducers.

e. Application of Infrared Radiation to Nondestructive Testing. Investigation of the possible use of infrared in nondestructive testing has continued. The mechanically scanned infrared imaging system that has been constructed has thus far not demonstrated sufficient temperature sensitivity for any testing applications. Several commercially available infrared radiometers are being investigated for purchase.

Some phosphor screens for an electronically scanned system have been fabricated. These screens were used in place of the transducer in the demountable ultrasonic imaging system. An image of the filament of an infrared lamp was produced by this method. The screens made thus far have been of poor quality.

The phosphor screen was made on soft glass which does not transmit in the infrared, so a high-temperature source was used to produce the image. When a good technique for producing uniform screens is developed, the screen will be made on an infrared-transmitting material so that the long-wavelength response can be investigated.

C. The Metallurgy Alpha-Gamma Hot Cell

Following the successful conclusion of a series of shielding and leakage tests, equipment for decanning and examination of capsules was set up in the gas cell and the cell was flooded with nitrogen. Four irradiated capsules containing cobalt-base control materials were introduced through the small gas interlock, and jacketed assemblies were removed from the NaK-filled capsules. At the beginning of the operation the gas cell nitrogen contained 0.8% O_2 and 1080 ppm H_2O .

The tests of the cell involved checking for weak areas in the concrete and steel shielding as well as cracks, which would permit in-leakage of air to contaminate the cell atmosphere with oxygen and moisture vapor. Shielding tests were made by monitoring the entire facility with Co^{60} . The leak tests were made by checking all welds and penetrations through the inner lining of the gas cell with a mass-spectrometer helium leak detector. Finally, leak-rate tests were made on the gas cell under operating conditions, and nitrogen-purge rates were determined.

The leak rate tests established a nominal in-leakage of less than 2.4 cc/sec at the design operating pressure of -1.3 cm WC. Although not

yet definitely established, in-leakage is believed to be primarily pure nitrogen from the pressurized seals provided at utility and equipment penetrations into the cell. Effective purging is possible with as little as 1.5 ℓ /sec nitrogen flow. However, about 24 hr is required to bring the 108,000- ℓ volume of the gas cell down to less than 1% oxygen with the higher purge rate of 7 ℓ /sec of nitrogen.

In the initial operations, the gas cell is being activated in a manner which will permit assessment and correction of some of the operational problems involved in maintaining a nitrogen atmosphere, and in making transfers of specimens and equipment into and out of a sealed cell. At the same time, however, final preparations are being made to activate the air cells in their role of supporting facilities for the operating gas cell.

D. Heat Engineering

1. Refractory Metal Test Loop

Difficulty has been experienced in procuring columbium alloy piping commercially for use in the instrumentation lines. The adequacy of the design of the bakeout heating system for the vacuum facility is being investigated.

2. Boiling Sodium Two-phase Flow Studies

The boiling sodium loop designed to investigate two-phase pressure losses and vapor volume fractions in a forced convection system is in initial operation. Calibrations of heat loss have been completed. The liquid-level gauges, which are used for differential pressure measurement and stability control, are being subjected to final tests in preparation for the introduction of sodium to the high-temperature portion of the loop.

E. Chemical Separations

1. Chemistry of Liquid Metals

a. Distillation of Liquid Metals. An additional run has been made in the large unit for distilling cadmium to determine the extent of entrainment of liquid cadmium in cadmium vapor (see Progress Report for August 1962, ANL-6610, p. 59). Prior to this latest run, a de-entrainment device for removal of entrained droplets from the cadmium vapor stream and an eddy-current induction probe for determination of the liquid level in the still pot had been installed in the distillation unit. The cadmium feed solution contained 0.3 w/o tin. Since tin is comparatively nonvolatile under the distillation conditions used, the concentration of tin in the distillate provides a measure of entrainment during still operation. Samples of the distillate are being analyzed for tin, and information concerning the

extent of entrainment will become available when the analyses are completed. The still was operated satisfactorily at a furnace control temperature of 570°C and distillation rates up to 80 kg/hr. Under these conditions, the liquid level in the still pot oscillated less than $\frac{1}{4}$ in. as indicated by readings of the liquid-level probe.

Studies (see Progress Report for July 1963, ANL-6764, p. 50) of the vaporization of mercury from a liquid pool without the formation of bubbles (nonturbulent vaporization) were continued. In the most recent work, a pool of mercury, $2\frac{5}{8}$ in. in diameter and 3 in. deep, was induction heated with 10-kc power. While the pool was heated at low pressure (<1 mm Hg) to effect nonturbulent vaporization, three vertical traverses of the pool centerline were made with a special thermocouple. In these traverses, steep temperature gradients of 11, 13, and 17°C through ~0.16-in.-thick surface layers were observed. The temperatures at the pool surface were 173, 176, and 181°C, respectively. In each traverse, the temperature of the mercury below the 0.16-in. surface layer was found to be almost constant. These results are similar to those obtained in earlier runs.

The heat source for the mercury pool was changed to an external cylindrical resistance heater, and vertical traverses of the pool centerline were then made at two vaporization rates. After this change there were also steep temperature gradients (of 11°C and 20°C) in a 0.17-in.-thick surface layer. Both the surface temperatures (160 and 164°C) and temperatures below the surface layer (171 and 184°C) were lower than with induction heating. Circulation patterns differed for the two types of heating.

b. Plutonium Recovery Process. Cadmium-zinc-magnesium alloys are being considered as process media for plutonium-bearing reactor fuels. The use of cadmium-rich alloys of these elements may be a means of lowering corrosion rates in stainless steel equipment and allowing operations at lower temperatures than those required in systems employing magnesium-zinc alloys. As a first step in the evaluation of the practicality of the ternary system, the solubility of uranium in the composition range of process interest is being determined. It has been found that at 600°C in cadmium solutions containing either 6 a/o zinc and 10 a/o magnesium or 13 a/o zinc and 10 a/o magnesium, the uranium solubility is greater than 1 a/o. The solubility of uranium in pure zinc is 0.05 a/o, and the solubility in pure cadmium is 1.02 a/o. Although the solubility of uranium in the mixed solvents thus far observed is about the same as that in pure cadmium, it is believed that the continuation of the investigation of the solubilities of uranium in the mixed solvents is worthwhile.

2. Fluidization and Volatility Separations Processes

a. Laboratory-scale Fluid-bed Fluorinations. Laboratory support work has been directed toward the establishment of optimum conditions for

the fluorination of mixtures of U_3O_8 and plutonium dioxide in a $1\frac{1}{2}$ -in.-diameter fluid-bed reactor. Fluorination experiments have been carried out in which selected fission product oxides were added to the mixture of U_3O_8 and PuO_2 in order to determine what effect the presence of the fission product oxides would have on the retention of plutonium by the alumina bed. The U_3O_8 - PuO_2 mixture contained about 0.4 w/o PuO_2 and 0.86 w/o of a mixture of 11 fission product oxides (La_2O_3 , CeO_2 , Pr_6O_{11} , Nd_2O_3 , Sm_2O_3 , Eu_2O_3 , Gd_2O_3 , Y_2O_3 , BaO , ZrO_2 , and MoO_3). Alumina was added to the resulting mixture prior to being fed to the fluid-bed reactor. In these experiments, the quantity of uranium and plutonium relative to the amount of alumina prior to fluorination is much less than would be used in plant operation and is about one-sixth of the amount used in previous boat experiments (see Progress Report for October 1962, ANL-6635, p. 43). The amount of uranium and plutonium retained by the alumina is used for comparing the results of various runs, since comparisons made on this basis are more equitable than comparisons which are based on percentages of uranium and plutonium that have been fluorinated.

The fluorination reaction sequence consists of two steps. The first step is a fluorination-feeding period (one hour) during which the feed material is continuously transported into a fluidized bed of alumina at $500^\circ C$ and simultaneously reacted with 20 v/o fluorine in nitrogen. The second fluorination step is carried out for a period of 5 or 15 hr at bed temperatures of 500 or $550^\circ C$, using recycled 100 percent fluorine.

Two experiments were made in which the fluorination reaction was terminated at the end of the fluorination-feeding period. In the first experiment, the feed charge did not contain added fission product oxides. The residual uranium and plutonium contents of the alumina were 0.043 and 0.063 w/o, respectively. In the second experiment, the feed charge did contain added fission product oxides. The residual uranium and plutonium contents of the alumina were 0.15 and 0.11 w/o, respectively. Two experiments were also made in which the recycle fluorination period following the fluorination-feeding period was carried out at a bed temperature of $550^\circ C$ for 5 hr for one experiment and for 15 hr for the other. The feed charge in these runs contained added fission product oxide. With a recycle fluorination period of 5 hr, the residual uranium and plutonium contents of the alumina were 0.017 and 0.038 w/o, respectively. With a recycle-fluorination period of 15 hr, the residual uranium and plutonium contents were 0.021 and 0.029 w/o, respectively.

The effect of using the same batch of fluid-bed alumina for three successive fluorinations of feed material (U_3O_8 , PuO_2 , fission product oxides, and alumina) on uranium and plutonium retention by the alumina was studied in one experiment. If the plutonium that accumulates during successive U_3O_8 - PuO_2 addition periods could be reduced to a low level by a final fluorination period at $550^\circ C$, then an overall reduction in the quantity

of plutonium retained by the alumina (which is discarded as waste) could be achieved. An economy in the use of alumina and in the quantity of waste to be handled would thus be achieved by this procedure. In each of the three fluorinations, a 5-hr recycle-fluorination step at a bed temperature of 500°C was used following the fluorination-feeding period. The results of this experiment showed that the residual uranium content of the alumina was low. However, the residual plutonium content of the alumina increased with each successive batch fluorination. The residual plutonium contents on the alumina were 0.057, 0.114, and 0.167 w/o. Tests are being planned to determine whether residual plutonium levels will continue to increase with successive batch fluorinations or reach a steady state.

b. Stress-corrosion-testing of Alloys of Aluminum and Titanium.

At the request of the Oak Ridge Diffusion Plant, samples of alloys of aluminum and of titanium were tested for stress corrosion in an atmosphere of plutonium hexafluoride. Four samples of 7075 aluminum alloy were stress-loaded to $61,000 \pm 600$ psi. Two samples of B-120 VCA titanium alloy were stress-loaded to $140,000 \pm 1,100$ psi. The stressed samples were placed in a quartz vessel, and the plutonium hexafluoride (at a pressure of 52 mm Hg) added. The temperature of the quartz vessel was held at 28°C for 15 hr, after which the temperature was increased to $65 \pm 3^\circ\text{C}$. The vessel is to be maintained at this temperature for the duration of the test (30 days). The two titanium alloy samples failed during the first 6 days of testing. After 21 days, the aluminum alloy samples were still intact.

c. Separation of Uranium from Aluminum-alloy Fuels by Chlorination and Fluorination Steps. Studies of the feasibility of using the fluid-bed chlorination-fluorination scheme for the recovery of uranium from uranium-aluminum alloy fuels were continued (see Progress Report for June 1963, ANL-6749, p. 46).

The effects of the following changes in experimental conditions on uranium recovery were investigated: (a) the use of less-expensive sintered alumina grain as an inert fluid-bed solid instead of fused alumina grain, (b) the presence of moisture (1100 ppm) in the hydrogen chloride feed, (c) the presence of hydrogen (20 v/o) in the feed hydrogen chloride, and (d) increase in the height of the packed-bed filter.

A run was carried out in a $1\frac{1}{2}$ -in.-diameter nickel reactor. The charge in the run consisted of three 57.3-g uranium-aluminum alloy subassemblies (5.2 w/o normal uranium), each subassembly consisting of five plates. The plates were 5 in. long by $\frac{7}{8}$ in. wide by $\frac{1}{20}$ in. thick, and were spaced $\frac{1}{20}$ in. apart. The reaction bed contained 320 g of sintered alumina grain (Tabular Alumina T-61; -40 +120 Mesh; Aluminum Company of America). The packed-bed filter contained 430 g of fused alumina grain (Alundum, Type 38; -14 +20 mesh; Norton Company).

The following reaction sequence was employed in the run: hydrochlorination, hydrofluorination, and fluorination. The hydrochlorination reaction was carried out while maintaining the fluid bed at a temperature of 250°C and the filter bed at 200°C. In the hydrochlorination period, two subassemblies were reacted with hydrogen chloride containing 1100 ppm water and 20 v/o hydrogen over a period of 3.3 hr, after which the third subassembly was charged to the fluid-bed reactor and reacted with hydrogen chloride containing 1100 ppm water (no hydrogen present) over a period of 4.2 hr. The hydrochlorination step was carried out at an average rate of 23 g of alloy per hour with three times the stoichiometric amount of hydrogen chloride. The hydrofluorination step (25 to 50 v/o hydrogen fluoride, remainder nitrogen) was conducted at a fluid-bed temperature of 350°C for a period of one hour. The fluorination period was conducted in two steps of 2 hr each in which the fluid-bed temperature was first maintained at 250°C and then maintained at 500°C. The fluorine concentration was gradually increased from 5 to 90 v/o (remainder nitrogen) during each period. During the hydrofluorination and fluorination reaction steps, the filter bed was maintained at the same temperature as the fluid bed.

The uranium recovered was 99.6 percent of the uranium in the initial charge (8.9 g). The uranium retained by the beds of alumina grain at the end of the reaction cycle was 0.3 percent. The loss through the packed-bed filter during hydrochlorination was 0.1 percent. The concentration of uranium retained by the sintered alumina grain in the fluid bed was 0.004 w/o and that retained by the fused alumina grain in the filter bed was 0.003 w/o.

The less-expensive, Tabular alumina T-61 did not cake or undergo attrition (less than 0.8% passed through 200 mesh at the end of the run). This material appears to be a satisfactory, inert bed material.

The use of hydrogen chloride containing water did not appear to affect either the overall hydrochlorination rate or the utilization efficiency of hydrogen chloride. The presence of hydrogen in the hydrogen chloride feed apparently did not affect the hydrochlorination rate or the uranium loss through the packed-bed filter. The uranium losses through the filter (collected in the condenser) corresponded to 5.0×10^{-5} g of uranium per gram of aluminum when 20 v/o hydrogen was used and 3.4×10^{-5} g uranium per gram of aluminum when no hydrogen was present.

d. Pilot-plant Facility for Recovery of Uranium from Enriched Uranium, Aluminum, and Zirconium Alloy Fuels. Shakedown runs have been started in the pilot plant installed to demonstrate the fluid-bed fluoride volatility process for the recovery of uranium from enriched uranium-alloy fuels (see Progress Report for May 1963, ANL-6739, p. 54). In initial experiments, the hydrochlorination and hydrofluorination reaction steps will be carried out with fuel assemblies composed only

of Zircaloy or aluminum (no uranium present). Later experiments will be made with assemblies containing uranium.

3. General Chemistry and Chemical Engineering

a. Conversion of Uranium Hexafluoride to Uranium Dioxide: Preparation of High-density Particles. The fluid-bed preparation of dense spheroidal uranium dioxide particles directly from uranium hexafluoride by the simultaneous reaction with hydrogen and steam continues to be investigated preparatory to a study which is planned for the conversion of mixtures of plutonium hexafluoride-uranium hexafluoride to the oxide. In current work, the feasibility of preparing small mesh size (~140 mesh) uranium dioxide particles was explored. The operating conditions used in the run for producing 140-mesh uranium dioxide particles were similar to those in previous runs with larger particles (see Progress Report for July 1963, ANL-6764, p. 56) except for a reduction in the superficial gas velocity (from 1.0 ft/sec to 0.50 to 0.75 ft/sec). A high proportion (over 50%) of dense, -140 +170 mesh uranium dioxide particles was produced from a starting bed of -170 +325 mesh uranium dioxide particles. The particle density of the as-produced uranium dioxide product was 9.8 g/cc (about 89% of theoretical density). This density value is the highest achieved in runs made at a reaction temperature of 650°C.

A brief study of an oxidation-reduction procedure for the preparation of dense uranium dioxide seed particles was also carried out. The runs were carried out with fluid beds of -40 +60 mesh uranium dioxide particles (a particle density of 8.8 g/cc). Mixtures of air and nitrogen were used as the oxidizing agent, and hydrogen was used as the reducing agent. The runs were carried out at 500°C and 650°C. At 650°C, fragmented material having a particle density of 7.2 g/cc was obtained. At 500°C, the particle density was 5.2 g/cc. Particle breakup was rapid and extensive after about one-half the stoichiometric amount of oxygen required for complete oxidation of the UO_2 to U_3O_8 was added; little particle breakup was noted before this point. Of the fragmented portion, about 50 percent was in the -325 mesh fraction. This fluid-bed method of preparing seed bed particles offers promise as an alternative to the present method of mechanical grinding.

Plans are being made to study the conversion of mixtures of uranium hexafluoride and plutonium hexafluoride to the oxide by reaction with steam and hydrogen in a fluid-bed system. This work will be carried out in a 2-in.-diameter reactor which is being fabricated and which will be installed in the engineering-scale plutonium handling facility.⁷

⁷Chemical Engineering Division Summary Report for Oct, Nov, Dec 1962, ANL-6648, p. 143.

b. Gas-phase Reaction for the Production of Plutonium Compounds. Sparging experiments with lead and aluminum have been conducted in the mockup reaction chamber (see Progress Report for August 1963, ANL-6780, p. 46). Revisions of the apparatus suggested by the results of the experiments have been made.

A gas-purification train has been installed in Glovebox PF-21. Sampling lines for gas analysis are now being connected to the purification train.

4. Calorimetry

a. High-temperature Enthalpy Calorimeter. Components of a calorimetric system to measure changes in enthalpy up to 1500°C are being tested. The calorimeter furnace has been cycled between 700-900°C and about 1300°C, being maintained at 1300°C for periods of 1 to 3 hr per day over a two-week period. During the course of this cycling the resistance of the furnace windings apparently stabilized so that the power required to maintain a given temperature of the furnace core was constant.

b. Fluorine Bomb Calorimetry. Exploratory combustions have established a suitable technique for burning tantalum diboride and niobium diboride in fluorine. A series of calorimetric combustions of niobium diboride in fluorine has been completed. Analysis of the thermochemical data for aluminum fluoride indicates that the heat of formation value of -356.5 kcal/mole (see Progress Report for September 1962, ANL-6619, p. 51) obtained by fluorine bomb calorimetry is too low and suggests that -358.0 ± 0.5 kcal/mole is a preferable value.

F. Plutonium Recycle Program

1. Hydrodynamics

The geometrically uniform active core (all fuel rods are 0.420 in. in diameter and have 4-ft active length) is divided into three radial fuel-composition zones: a central plutonium zone, an enriched uranium shim zone, and an outer natural uranium zone (see Progress Report for June 1963, ANL-6749, p. 49). In performing the power-void iteration calculations to reconcile the physics and hydrodynamics results, the core is divided into four radial regions (the shim zone is divided into two regions) and three axial regions per fuel zone. Using the recently revised "CHOPPED" IBM-704 code for hydrodynamics (RE-293), the iterations have proceeded at a considerably faster pace than heretofore possible. For the aforementioned fixed core geometry, PDQ source points and integrated average regional power densities may now be used as direct input to the "CHOPPED" program. The resultant void-fraction profiles are plotted and the graphically averaged void values are used in the succeeding PDQ calculation.

Curves of void fraction per region vs. total reactor power have been generated for reactor powers of 20, 30, 40, and 50 Mw, for zero and 0.6 percent burnup. Separate iterations must be run for each level power and each burnup value, since, in general, the power fraction per radial fuel zone does not remain constant. Calculations are in progress to verify the accuracy of the assumed subcooling and riser void values necessary for input to the hydrodynamics program.

2. Plutonium Recycle Fuel

The central zone of the experimental core will contain 36 plutonium fuel elements with a fuel composition of $U^{238}O_2$ -1.5 w/o PuO_2 . This will be surrounded with a double row of 60 shim-zone elements fueled with UO_2 enriched with 6 w/o U^{235} . Shim-zone fuel elements will contain the burnable poisons Eu_2O_3 and Sm_2O_3 to extend the life of the fuel and limit the range of excess reactivity present over the period of irradiation. A third zone of 58 blanket elements containing natural UO_2 will surround the shim zone. All enriched and natural uranium fueled elements are referred to as the Plutonium Recycle Driver Fuel.

The fuel for all rods is contained in 0.945-cm-ID x 0.056-cm-wall Zircaloy-2 tubing. A fuel element contains 36 fuel rods in a square lattice. The 36 fuel rods are supported and positioned by support grids and spaced by a center spacer grid. The fuel rods and grids are contained within and fastened to a square Zircaloy-2 fuel-element can. A stainless steel end fitting completes the fuel element.

Plutonium-bearing fuel rods for the Plutonium Recycle Experiment are being fabricated by General Electric Hanford. The plutonium fuel rods will be assembled into fuel elements at ANL. All driver fuel, components for assembly of plutonium fuel elements, and components for future replacement of 28 driver shim elements are being procured from a commercial fuel contractor by ANL.

Hanford has completed procurement of fuel-rod components. Zircaloy-2 tubing fabricated at Harvey Aluminum for Hanford was inspected by Hanford. Comparison of nondestructive testing procedures for optimum inspection of this tubing was a joint effort between Hanford and ANL (see June and July Reactor Development Program Progress Reports, ANL-6749 and ANL-6764).

Hanford is preparing all PuO_2 - UO_2 fuel by the Dynapak process. This is a high-temperature, high-pressure impacting process which produces a uniform dispersion of PuO_2 in UO_2 . Dynapaked material is being crushed, screened, and loaded into the fuel tubes by vibratory compaction. This process is being used for all plutonium fuel in place of the incremental loading and vibratory compaction which was originally proposed.

Plutonium fuel rods will be completed and available for the physics tests which are to be performed at Hanford beginning the third quarter of FY 1964.

A contract is being negotiated by the Laboratory with United Nuclear Corporation for procurement of the driver fuel. United Nuclear has agreed to meet the technical requirements and specifications to the Laboratory's satisfaction. During the course of negotiation some questions were raised regarding the degree of homogeneity which could be maintained for the rare earth oxide in UO_2 in the shim elements. United Nuclear Corporation's Chemical Division does not have facilities for preparation of the rare earth oxide and UO_2 by coprecipitation. Attempts to blend small quantities of rare earth oxide with ceramic-grade UO_2 by mechanical mixing of dry powders showed that a wider distribution of rare earth was obtained in the UO_2 than was desired. Other methods of adding the rare earths will be tried:

- (a) preparation of a master blend of high-fired rare earth oxides with UO_2 , blended with ceramic grade UO_2 ;
- (b) preparation of a master blend of rare earth oxide and UO_2 by the Dynapak high-energy-rate forming process followed by grinding, screening, and blending with ceramic-grade UO_2 .

Pellets prepared from the above materials will be fabricated and analyzed for rare earth homogeneity. In the event that neither one of these procedures will produce satisfactory dispersions, an alternative procedure of blending rare earth oxides and UO_2 powders in small quantities and random stacking of pellets for loading into fuel rods will be used. This alternative method is admittedly more expensive and every effort will be made by the contractor to avoid using this procedure.

3. Plutonium Recycle Control Rods

A new set of control rods will be fabricated to replace existing control rods in the EBWR reactor. The existing control rods have riveted transition sections between the boron-stainless absorber section and the Zircaloy-2 follower section. A number of Inconel rivets in these transition sections have failed in service. It has been determined that a shear stress exists in these rivets due to differential expansion between the Zircaloy and the stainless steel. Metallographic examination of failed rivets has indicated that stress corrosion may also be a factor in the failure of these rivets. Irradiation damage may have contributed to the stress corrosion failure.

The control rods for replacement have been designed with changes to eliminate riveted joints. New drawings are being prepared incorporating the necessary design changes.

Along with the change in mechanical design, the new control rods will contain highly enriched ($\sim 90\%$) B^{10} in place of natural boron. The total boron content will remain at 2 w/o. The use of enriched rather than natural boron will provide some additional rod worth due to increased epithermal absorptions.

A total of eight control rods containing boron are required. One central oscillator rod with hafnium as the absorber material will be replaced with a new hafnium rod.

All boron stainless steel required for control rod fabrication will be procured by the Laboratory from commercial suppliers. Specifications are completed for the boron-stainless steel and invitations are being prepared for bids on the fabrication of the steel. The boron-stainless steel for the eight boron rods and hafnium for the oscillator rod will be furnished by the Laboratory to commercial suppliers who will fabricate the control rods. Specifications will be prepared for procurement of the control rod fabrication as soon as new control rod drawings are released.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Facility Design

Based on heat transfer studies performed in late 1962 at the Argonne National Laboratory to evaluate the feasibility of increasing power to 240 Mw by an extension of existing technology, it has been decided to upgrade the original design to the higher power. An ultimate goal of 1×10^{16} nv thermal in the internal thermal column (corresponding to about 2×10^{15} nv in the experimental beam tubes) has been established. By taking advantage of certain factors such as controlled local boiling in the reference reactor core, physics and heat transfer evaluations have indicated the feasibility of achieving the ultimate flux with a reactor power of 240 Mw.

Although the extension of existing reactor technology represented by controlled local boiling operation must be supported by appropriate research and development, primarily in the heat transfer and fuel development areas, the likelihood of success is sufficiently high to proceed with revision of the facility design for the higher power level.

A revised process system design was prepared by the Laboratory, and the facility building design and arrangements were modified for the increased power by Shaw Metz and Associates (see Progress Report for May 1963, ANL-6739, p. 56). Since that time additional changes in building arrangement and equipment location have been made. In August, the Laboratory contracted with United Nuclear Corporation for the preparation of a Revised Preliminary Cost Study of the upgraded reactor and supporting facilities. Shaw Metz and Associates participated in the preparation of the cost estimate under separate subcontract to the Laboratory. The cost study was completed and submitted to the Laboratory in September, 1963.

2. Core Physics

Calculations of the flux distribution for the 40-40 (0.040-in.-thick fuel plates and 0.040-in. coolant-channel spacing) core at 240 Mw have been made; these show a peak thermal flux of 10^{16} nv at the center, 2.5×10^{15} nv in the reflector, and an initial excess k above xenon and samarium of 15%. The flux gradient in the reflector varies from 1%/cm for thermal neutrons at the thermal peak to 10%/cm for 100-ev neutrons at their "peak" next to the core.

Flow studies of the rectangular channels for the 40-40 core indicate that some reinforcement is required for the fuel plates. To prevent excessive deflections a longitudinal insert can be used. Preliminary

calculations show that inserting 0.040 x 0.040-in. spacers and removing fuel from beneath the spacers causes a loss of 0.8% k_{ex} and a small shift in flux. Spacers with fuel beneath them are also being considered; however, in view of their small size, additional work will be required to determine feasibility.

Analysis of heat transfer from a hot channel next to a control rod inserted half way indicates that tests will be necessary to establish the burnout criterion for these channels, since the computed departure of the heat flux from nucleate boiling is within the uncertainty of critical heat flux correlations.

3. Critical Experiment

A cost-plus-fixed-fee contract is being negotiated for the procurement of fuel. It is anticipated that the earliest delivery (in part) will probably be no sooner than March 1964.

4. Studies of Reactor Control

During September the calculations necessary for determination of the core transfer functions at 240 Mw were completed, and a stability estimate was made. Also, since there have been some questions regarding flow coastdown characteristics of the primary loop, calculations pertaining to this phenomenon have been made.

It appears that the analog program used to study core transients around 100 Mw may be useful in a preliminary study of transients around 240 Mw wherein boiling occurs in small core regions for substantial reactivity insertion. It is planned to perform analog runs on this basis and concurrently to draw up specifications for a digital program which will better detail steam distribution when the steady-state boiling power threshold is below or near reference power. It is anticipated that both of these tasks may be brought near completion in the coming month.

5. Fuel and Core Design

Two reduced-size, simulated fuel-assembly test sections FTS-2A and FTS-2B were subjected to isothermal (100°F) conditions of water flow in the range of interest to AARR.

FTS-2A and FTS-2B consisted of 16 Type 304 stainless steel flat plates, 0.0375 in. thick, 2.315 in. wide, and 7.128 in. long that were spaced 0.0405 in. apart to form flow channels. The plates were held together by a dovetail key arrangement that was proposed for AARR fuel assemblies. The basic difference between the test sections was that FTS-2B had inlet and exit combs for increasing the plate rigidity.

The tests resulted in the following conclusions: a) the dovetail keyed type of assembly as presently devised is inadequate for AARR since it allowed excessive amplitude and vibration in the region of 50 to 55 ft/sec even when inlet and outlet support combs are used and the inlet edges facing the flow are rounded to decrease flow resistance; b) the maximum amplitude of the plate deflection occurred in the proximity (~ 0.2 the key spacing) of the downstream keys midway across the width. (Intuitively, the maximum deflection would be expected to occur at the transverse and longitudinal midpoints between the keys.)

Future tests will be conducted with the test sections held vertically. The flow will be directed downward because in the tests described above, air bubbles remained stationary, bridging the coolant gap between plate surfaces even though the flow velocity was as high as 65 ft/sec. Although the bubbles were few in number and did not cause the excessive vibrations, it is important to know what their behavior will be in a vertical, closely spaced fuel assembly.

6. Design of Experimental Facility Beam Tube

A subcontract has been negotiated with the Franklin Institute to perform stress analyses of three selected beam tubes for AARR in the region of the core utilizing two power levels (100 Mw and 240 Mw) and two pressures (500 psig and 800 psig). This study will provide a complete pressure stress and thermal stress analysis for determining optimum thickness of the cylinder and the hemispherical end of the three specified beam tubes. Discontinuity stresses, temperature distribution, and flow and cooling requirements will also be determined. It is expected that the work will be completed by the end of November, 1963.

7. Shielding

The upgrading of the reactor power level, and consequent rearrangement of equipment and buildings, has necessitated a review of the shielding requirements. Dose-rate criteria have been established for most of the areas, and studies are in progress to determine the new shielding requirements. The following shielding calculations have been completed to date; a) Hot Cell (based on a spent fuel assembly, which is considered the most intense source to be placed in this area); b) wall separating auxiliary coolant system cubicle from Office and Laboratory Building; c) Fluid Waste Tank (based on 1% of the fission products being uniformly dispersed in 25,000 gal of water).

B. Magnetohydrodynamics

A preliminary experiment has been performed to determine the feasibility of generating ac power by MHD methods. Figure 12 shows a

schematic of the experiment which was performed at room temperature whereby alternate slugs of liquid metal (NaK) and nitrogen gas were allowed to flow at high velocity through a magnetic field. The object of the experiment was to demonstrate that slugs of liquid could be propelled through a duct and remain intact through the generator. In addition it was also possible to examine the wave form of the voltage and current pulses, and to determine the pulse shape and reproducibility. Figure 13 shows the oscillograph tracing of the secondary voltage with a load (a 4-w, 115-v lamp). This tracing shows operation at 8 cycles/sec.

Figure 12. A Schematic of Apparatus Used in an Experiment to Generate A-C Current by Means of a Liquid Metal MHD Generator

Figure 13

The wave form of the A-C output from the liquid metal MHD generator was obtained from a recording oscilloscope. The generator was connected to a transformer having a primary-to-secondary turn ratio of 1:1600. A 4-w, 115-v lamp was used to load the secondary transformer winding.

The valving for the slug formation was rather crude and could not perform at the higher frequency. Fabrication of refined apparatus is now in progress. Experiments are planned to examine and improve the mechanism of the slug formation, acceleration, efficiency of power, generation, and electrical pulse output.

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal Oxidation and Ignition Studies

a. Isothermal Oxidation of Plutonium-Aluminum Alloy. In previous studies (see Progress Report for July 1963, ANL-6764, p. 64), it was found that the addition of 2 a/o aluminum to plutonium increased the ignition temperature of plutonium. To gain a better insight into the reasons for this phenomenon, studies of the isothermal oxidation of plutonium-2 a/o aluminum alloy were undertaken. Over the temperature range from 150 to 588°C, the oxidation behavior of the alloy in air or in oxygen was, in general, similar to the behavior of pure plutonium (see Progress Report for March 1963, ANL-6705, p. 63). In both systems, the oxidation followed a parabolic rate law which later in the oxidation became linear.

In the temperature region above 420°C (high-temperature regime), the parabolic rate constants for the alloy were found to be very similar to those for unalloyed plutonium. However, the linear rate constants for the alloy were lower by about a factor of two than those for unalloyed plutonium. The activation energies for the parabolic and linear oxidation processes in the high-temperature regime were the same for both systems, being about 32 kcal/mole for the parabolic process and 29 kcal/mole for the linear process.

In the temperature region below 420°C (low-temperature regime), the rate constants for both the parabolic and linear oxidation processes were much lower for the plutonium-aluminum alloy than for the unalloyed plutonium. This accounts for the higher ignition temperatures of the alloy. The activation energies for the two systems differed for both the parabolic and linear oxidation processes: 14 kcal/mole for the parabolic process and 13 kcal/mole for the linear process. In the case of the unalloyed plutonium, the activation energy was 32 kcal/mole for the parabolic process and 16 kcal/mole for the linear process.

b. Oxidation of High-purity Plutonium. A brief study of the oxidation of high-purity plutonium has been initiated to determine if the normal impurities found in reactor-grade plutonium affected the ignition temperature of the plutonium. The impurity content of the high-purity plutonium sample was lower by an order of magnitude than that of the reactor-grade plutonium used in previous studies: 99.84 a/o as compared with a purity of 98.6 a/o for typical reactor-grade specimens of plutonium.

Preliminary results indicate that the oxidation behavior of the high-purity sample is the same as that of the reactor-grade plutonium at

240 and 300°C, but that the behaviors of the two types of plutonium may be different below 200°C. Additional experiments are planned in which rate data will be obtained for the oxidation at temperatures below 200°C and above 420°C (the approximate transition temperature to the high-temperature regime).

c. Thermal Conductivity of Powders. A method for the measurement of the thermal conductivities of powders is being developed. It is anticipated that the studies of the effects of particle size, temperature, and degree of oxidation on thermal conductivity will shed light on the mechanism of ignition of metal powders. The method that is being developed utilizes a line source of heat which is located along the centerline of a cylindrical, stainless steel container, of $\frac{3}{4}$ -in. ID by 2 in. high. Determinations of thermal conductivities are made by filling the container with the test material, and measuring the rate of heat production and the temperature of the line source relative to the temperature outside the container. The line source consists of a fine constantan heating wire which, along with iron-constantan thermocouples, is placed inside a stainless steel hypodermic tube. Two line sources have been constructed, one with 62-mil-diameter hypodermic tubing and the other with 25-mil-diameter tubing.

To check the cell and associated equipment, tests were carried out with Lucite and paraffin using the 62-mil-diameter line source. The measured values of the thermal conductivities of the Lucite and paraffin samples agreed within 5 percent with the literature values. The measurement of the thermal conductivity of water using the larger line source was not feasible because of convective effects. It is anticipated that these effects will be greatly diminished when the 25-mil-diameter line source is used.

d. Aluminum- U_3O_8 Reaction. Studies of the aluminum- U_3O_8 thermite reaction were continued (see Progress Report for July 1963, ANL-6764, p. 65). The re-examination of this problem was prompted by a recent publication⁸ in which it was reported that a thermite reaction occurred in aluminum- U_3O_8 cermet. Ignition of mixtures containing more than 75 w/o U_3O_8 were reported, and calculations were made which showed that considerable energy is potentially available even at 50 w/o U_3O_8 .

Three additional experiments were carried out at Argonne in which cold-pressed compacts of aluminum with 45, 60, and 75 w/o U_3O_8 were used. Each of the compacts contained 3 w/o water. (Fleming and Johnson's experimental samples contained some water.) In each case, the temperature-time trace obtained by heating the compacts at a rate of 25°C/min was identical with the trace obtained in previous experiments with dry compacts with corresponding aluminum: U_3O_8 weight ratios. Calculations of the extent of reaction achieved in the three runs await completion of analytical studies.

⁸J. D. Fleming and J. W. Johnson, *Nucleonics* 21(5), 84 (1963).

2. Metal-Water Studies

a. Stainless Steel-Steam Reaction. Studies of the reaction between stainless steel and steam were continued (see Progress Report for May 1963, ANL-6739, p. 62). The method that is being used depends upon the measurement of the hydrogen evolved during the course of the reaction. At 1300°C, it was found that the rate of hydrogen evolution in the early stages of the reaction depends upon the time for heatup, carried out in the presence of steam, required to achieve the test temperature. Thus, considerably higher rates of hydrogen evolution are obtained in the first 15 min of the reaction, when the heatup time is 3 min, than are obtained in runs in which the heatup time is 4 to 5 min. After 15 to 40 min, the rates of hydrogen evolution for all runs become the same. A further study of the effect of heatup time on the rate of reaction is being made.

b. Laser-beam Heating Applied to Metal-Water Reactions. Research is continuing on the application of laser beam heating to the metal-water reaction. High-speed motion pictures, which were taken during recent experiments, have shown that small discs of zirconium foil (of 2-mm diameter by 0.13 mm thick) submerged in water can cause a sharp upward motion of a water column 2 cm high and 1.5 cm in diameter when the zirconium disc is heated to incandescence in about one msec. The velocity of the upward ejection of the metal-water system is estimated to be about 250 cm/sec. Studies are being made to determine the conditions required for the ejection of the water and to determine the temperatures attained by the metal. Microanalytical apparatus for the measurement of the hydrogen generated by the metal-water reaction during experiments in closed reaction cells has been constructed and tested.

B. Fast Reactor Safety Studies

Fast power reactor designs require sufficient fuel that a super-prompt-critical assembly could be produced if the core were rearranged into a sufficiently compact mass. Hence, a class of "meltdown" accidents may be postulated in which some operating abnormality occurs. The core undergoes some degree of failure (or meltdown), and the fuel moves into a more compact assembly which produces a destructive burst of nuclear power. However, the interplay of mechanisms affecting fuel element failure and material movement is complex and not well-understood. Consequently, Argonne is engaged in a program, centering on experiments conducted in the TREAT reactor, to study the phenomena of meltdown and assist the safety analyst in assessing potential core safety problems.

1. In-pile Experiments of Pre-irradiated Metallic Elements

Experiments are being performed in TREAT with irradiated EBR-II Mark-I and Fermi Core-A samples in order to study effects of fission gas

inventories on meltdown. Previous results obtained with opaque capsules⁹ indicated sufficient expansion of the fuel upon melting to provide significant limitation on the degree of compaction possible for the meltdown products, at least in the early stages of a meltdown incident. However, these results are based on postmortem inspections only and are not necessarily representative of meltdown product densities during the failure and expulsion from the cladding.

Four special transparent capsules, designed for remote assembly, have been prepared in the caves for photographic experiments on meltdown of previously irradiated specimens. Two capsules contain single EBR-II elements and two half-length Fermi Core-A samples. The capsules have been loaded into their shipping coffins in preparation for transportation to TREAT for transient experiments.

2. Analysis for Coherence of Failure in Reactor

In order to investigate the possible use of meltdown information in safety analyses, calculations have been made of three hypothetical accidents for an EBR-II-like reactor. The detailed specifications for the three cases are somewhat artificial: however, they do present an interesting range of conditions, running from a case in which the time scale for fuel-element failure is determined by the time for penetration of fuel-element cladding by molten fuel, to a particularly extreme case in which the time of movement of meltdown products is essentially the time during which the fuel is expelled from a single element.

In the event of an accident in a fast reactor leading to fuel-element failure, the distribution in time of meltdown of different fuel elements determines the rate of reactivity change. In general, the reactivity change could be either negative or positive. Since these changes are not well understood, it will be assumed here that the changes are positive, and lack of coherence in failure times is desirable.

a. Case 1. Failure of Pumping Power. The EBR-II reference core consists of 61 subassemblies, each of which contains 91 fuel elements. The subassemblies are grouped into rows, depending on location with respect to the core centerline. Sodium coolant enters the bottom of each subassembly through an orifice, which is preset to vary the flow rate from row to row to reduce differences in coolant outlet temperature. Inside a subassembly, elements can be classed as (1) internal pins (which have an effective coolant cross-section area of 0.110 cm^2) and (2) peripheral pins (of 0.180-cm^2 area). The coolant flow rate decay curve used to describe the effects of pumping power failure has been given previously.¹⁰ Reactor power is assumed to remain constant throughout the flow transient at design EBR-II

⁹Progress Report for July 1963, ANL-6764, p. 65.

¹⁰EBR-II Hazards Report, ANL-5719 (1957).

level. Five cases were studied by means of the IBM-704 "ARGUS" transient heat transfer code:

- (1) central row, internal element;
- (2) central row, peripheral element;
- (3) fourth row, internal element;
- (4) fourth row, peripheral element 1 (nearest core center);
- (5) fourth row, peripheral element 2 (farthest from core center).

In the EBR-II reference design, the central and fourth rows are those of maximum power and maximum coolant outlet temperature, respectively. On the basis of analysis of meltdown experiment results, a fuel-cladding interface temperature of 1015°C is taken as the threshold for element failure by cladding dissolution, and a temperature of 1200°C is taken to be the threshold for failure due to bursting of the cladding. Penetration time for penetration of the cladding by molten fuel is assumed to be of the order of one second. Calculations show that the time spread within subassemblies is larger than between subassemblies in rows 1 and 4. Failure of one internal element can "trigger" failure of a nearby peripheral element (or vice versa) by means of molten fuel-cladding eutectic* from the failed pin eating its way into the cladding of the still intact neighbor. Hence, the time scale for element failures in this hypothetical accident is dominated by the time of cladding penetration. It would be of the order of 2 sec for each of the two subassembly locations, and of the order of 3.6 sec for all the elements in both subassembly rows.

b. Case 2. Blocked Orifice. For another set of problems, it was assumed that the flow rate through the subassembly decreases linearly in one second to 1% of design flow because of an unspecified orifice blockage. Reactor power is assumed to be constant. The assumption of constant reactor power is not as extreme in this case as in case 1, since the blocked-orifice incident is basically a single subassembly event, i.e., the negative reactivity change caused by temperature rise is smaller by almost two orders of magnitude than for a temperature excursion for the reactor as a whole. Calculated maximum fuel-cladding interface temperatures were obtained as a function of time for three element locations:

- (1) central row, internal element;
- (2) fourth row, peripheral element (nearest to core center);
- (3) fourth row, internal element.

For the fourth row subassembly, a spread of about 0.12 sec occurs in time of attaining the failure threshold, and this time should be combined with the duration of fuel movement, which is estimated to be of the same order of magnitude. In the incredible event of complete blockage of coolant at the core inlet, these cases imply a time spread of about 0.28 sec, which must

*The low-melting iron-uranium eutectic occurs at 725°C.

be combined with the duration of fuel movement for one element to obtain an estimate of the total duration of reactivity input. Inclusion of the central row peripheral elements and fourth row outermost peripheral elements would increase the spread somewhat but not change the order of magnitude of time for reactivity change.

c. Case 3. Fuel Subassembly Dropped during Loading. An extreme hypothetical loading accident was examined as case 3. It is assumed that the reactor is critical at low power, and that, at the worst possible position, the central fuel subassembly is dropped so as to fall freely into position in the core. Reactor feedback is estimated to be essentially that for adiabatic heating of the fuel, because of the extremely short duration of the excursion. Reactor neutron density and time-integrated neutron density were calculated by means of the IBM 704 kinetics code RP 129 H. In this case, the reactor becomes prompt critical at about 194 msec, within about 5.5 msec the average fuel temperature has risen to 1200°C, and the rate of rise at this time is about 160°C/msec. Taking into account the radial variation in subassembly power, the overall time spread in failure is estimated to be about 4 msec. Thus, the time duration of reactivity change would be essentially that for fuel movement of one element in case 3.

3. Analysis of Pressure Surges in Fast Reactor Coolant Channels

Development of techniques for calculation of the peak magnitude and shape of transient coolant pressure pulses due to temperature excursions in fast reactor coolant channels is important for the following reasons:

- (a) Pressure predictions are needed to specify design criteria for experimental meltdown loops and instrumentation.
- (b) Pressure surges during meltdown can affect the rate and direction of movement of meltdown products.
- (c) In core designs having positive sodium void coefficients, the transient expulsion of sodium coolant is a potential source of significant reactivity addition. Hence, expulsion rates must be known to determine the severity of hypothetical loss-of-coolant accidents.

An analytic model for a boiling coolant pressure pulse due to a given heat input is again being studied at length. In previous work done on this problem the solution to the final differential equation seemed to be the most formidable obstacle. This equation has been solved on the LGP-30. Sample problems have been run and the results appear consistent. A similar program is now being prepared for the IBM-704. Studies are underway on the scaling laws for this model, and on the relative magnitudes of errors introduced by the simplifying assumptions made in developing the model.

The first trial problem run with the LGP-30 was an application of the model to the small TREAT sodium loop. The data are based on an EBR-II, 3% enriched pin exposed in the loop to a TREAT constant-power transient of 157 Mw. Further, all the power generated by the fuel is considered to be entering the sodium. The pressure-time relation after coolant reaches saturation is shown in Figure 14. The maximum pressure generated by the vaporization of the sodium is approximately 8 atm, occurring at 56.6 msec after initial boiling. Preliminary calculations indicate that peak pressure is approximately proportional to $(\text{power})^{2/3}$. Thus, this phenomenon is not expected to present a hazard to package loop operation.

Figure 14

Calculated Pressure in Small TREAT Test Loop as a Function of Time after the Sodium Coolant Reaches Saturation Temperature

C. TREAT

1. Small TREAT Loop

The fabrication of the first loop is about complete. An EBR-II element with a cast-in Ta-Mo thermocouple has been prepared for testing in this loop and will be used in the first series of experiments.

The fabrication of the three other loops will take approximately another three months after the first loop is completed.

2. Large TREAT Loop

A preliminary report on the results of the thermal stress analysis made at the Franklin Institute has been received. The conditions of analysis were based upon the most severe tests to be conducted in the reactor. Two significant facts are noted in the conclusions of the report. The first

conclusion states that a thermal stress problem does not exist in the settling tank. The analysis indicates that the hot sodium slug which is ejected in a transient will cool sufficiently as it moves from the test section and, therefore, does not introduce thermal stresses beyond the test section. The second conclusion notes that, although significant stresses do develop in the test section, they do not reach destructive intensities. Based upon the preliminary results, the study has shown the design of the entire loop to be conservative and should be adequate for all of the planned experiments.

3. Experiments in Progress

Cylindrical, jacketed uranium oxide fuel elements were subjected to a series of transient irradiations to determine the thermal time constant of thermocouples contained within them. Such measurements are useful in determining the effective conductivity of these fuel elements.

A series of transient heating experiments on prototype Hallam Nuclear Power Facility (HNPF) uranium carbide fuel was in progress at the end of the month.

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