ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

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Albert V. Crewe, Laboratory Director

<u>Division</u>	<u>Director</u>
Chemical Engineering	S . Lawroski
Idaho	M. Novick
Metallurgy	F. G. Foote
Reactor Engineering	B. I. Spinrad
Remote Control	R. C. Goertz

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Report coordinated by R. M. Adams

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FOREWORD

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

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

The last six reports issued
in this series are:

November 1961	ANL-6473
December 1961	ANL-6485
January 1962	ANL-6509
February 1962	ANL-6525
March 1962	ANL-6544
April 1962	ANL-6565

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I. WATER COOLED REACTORS

A. General Research and Development

1. Heat Engineering

A study of the spectral shift reactor concept which is achieved by introducing voids in the downcomer of a boiling water reactor was noted in ANL-6565 (Progress Report April, 1962). For the assumed conditions of operation in an EBWR type reactor, calculations show that the coolant velocity in a system having zero void fractions in the downcomer and inlet plenum and a void fraction of 0.4 in the core exit fluid is 30% lower than the coolant velocity in a system having a void fraction of 0.2 in both the downcomer and in the inlet plenum and a void fraction of 0.6 in the core exit. The reduced coolant velocity results in a decrease of 15% to 20% in the gross power since some power flattening is achieved.

2. Instrumentation

Evaluation of two ultrasonic liquid level transducers was continued.

One unit was found (April Progress Report, ANL-6565) to have a questionable weld. X-ray examination of the weld showed that it was made so poorly that no conclusions could be drawn concerning its integrity. In an attempt to perform a hydrostatic test it was discovered that some internal mechanical failure, possibly resulting from temperature cycling, made the unit inoperative. No further tests are planned on this unit.

The second unit was found to perform satisfactorily at room temperature, although the resonant frequency was considerably different from that reported by the manufacturer. The unit also performed satisfactorily as a level alarm at 1500 psig and room temperature, but failed to operate at 500°F and 666 psig. Further tests will be performed in an effort to discover the cause of failure.

B. EBWR

1. Evaluation of Fissionable Material in EBWR

In connection with the IAEA inspection of EBWR, calculations were made to estimate the amounts of U^{235} , U^{238} , and plutonium present in the EBWR Core 1A, and in the reactor building as of May 31, 1962. These amounts are listed below:

	In the Core (kg)	In the Storage Pit	Total
U^{235}	90.390	3.180	93.570
U^{238}	5844.2	136.3	5680.5
Plutonium	7.816	0.076	7.892

2. Control Rod Calibration

A comparison of calibration results was made between the original system equipped with stainless steel fueled followers and the present system using Zircaloy followers. The comparison was based upon bank control, i.e., where all rods are moved together. The results indicate that the control rods with Zircaloy followers have about 0.4% more reactivity (ρ) when moved from 10 to 22 in., the distance being measured from the bottom of the core. The greater worth of the rods with the Zircaloy followers was, of course, anticipated.

3. Void Coefficient Measurements in EBWR

Additional measurements were made of the void coefficient. Four "void simulators" were used in four enriched thin elements to produce approximately 70% uniform voids. The void simulators provide for the substitution of air for water in the water channels of the fuel elements. The data are now being analyzed.

4. Core Change

The final loading change prior to initiating the next series of high power runs was completed. The change consisted of replacing one enriched (1.44%) thin fuel assembly with one enriched (1.44%) thick instrumented fuel assembly. The assembly is equipped with two turbine-type flow meters, two miniature ionization chambers, and thermocouples. This change increased the reactivity by approximately 0.11% ρ .

5. EBWR Transfer Functions

Transfer function measurements on EBWR were continued and performed under the following conditions:

- (1) 10 Mw, 300 psig, lower feedwater ring, and boric acid concentration about 4 gm/gal.
- (2) 10 and 20 Mw, 600 psig, upper feedwater ring, without boric acid.
- (3) 10 and 20 Mw, 300 psig, upper feedwater ring, without boric acid.

The measurements were made using the rod oscillator at the same 32 frequencies selected for last month's experiments from 0.0012 to 9 cycles per second. Data were recorded with the null analyzer, analog sine-cosine cross-correlator, and digital data system. Transient response records and noise records were made under each new condition of power and pressure. The RMS value of the reactor and noise power density spectrum were measured using an analog computer.

The last experiment in this test series used discrete-interval binary noise to simulate a statistical reactivity input. A 100-point switch, driven at constant velocity, was wired to give a 97-increment binary signal. The selection of whether the rod is to be up or down during a discrete time interval was based on the synthesis of a binary noise sample by using quadratic residues. The quadratic residues can be found from $q_n = n^2 - MN$ where M is an integer such that $MN < n^2 < (M + 1)N$ and N is a prime number. The set of quadratic residues q_n , $1 < n < (N - 1)/2$, give the locations in the sequence of one state of the binary digit. The control rod position and neutron flux were recorded with the digital data system. The IBM-704 digital computer was used to calculate the transfer function by cross-correlating the reactivity input and neutron flux output. The results of this experiment indicated that it would be desirable to have a longer binary record. It is possible to use a repetitive input signal, but the fundamental record must be a prime number. An alternate method of generating a discrete interval binary noise signal was developed. A square wave generator is used to determine the time per interval, and a random noise generator is used to determine the state of the rod (up or down) during the interval. The new discrete interval binary noise generator can generate a signal of any length, and the time per interval is easily selected by setting the square wave period. Preliminary results, which look promising, have been obtained by using the analog computer to simulate various transfer functions. These initial investigations were made under ideal conditions without noise. The work will be extended to investigate the effect of various signal to noise ratios on the accuracy of the transfer function measurement.

The transfer function data have been used to investigate reactor stability at 10 and 20 Mw at 600 psig without boric acid. The results indicate that the gain margin has been improved by six decibels and the phase margin has improved by 35 degrees as compared to the data obtained with the old 4-ft core. The new values of gain and phase margin, if extrapolated, indicate that instability occurs at a power level greater than 66 Mw. An estimate of an unstable power level will be made after additional data are obtained at higher power levels.

The inlet liquid velocity to power and exit liquid velocity to power transfer functions have been calculated and plotted from data recorded with the digital data system. The inlet liquid velocity and exit liquid velocity and exit liquid velocity signals are obtained from the turbine flowmeters installed on instrumented fuel assembly No. 2 located in core position No. 101-266.

C. BORAX-V

1. Operations and Experiments

Room temperature zero power experiments on the boiling core were completed in May. After completion of flux wire irradiations, the boric acid solution was drained from the reactor vessel and a flux wire irradiation was made with clean water to obtain information for comparison with boric acid solution in the system. Representative radial and axial traverses were obtained. Radial traverses were also made within the control rod region. As expected, the ratio between maximum and minimum values of thermal flux in a clean assembly are slightly greater than with boric acid.

Analysis of the fine flux plots has been completed. The fuel assembly peak-to-average power for the core with boric acid is 1.31, and the fuel rod peak-to-average power is 1.87. The 1.87 value compares favorably with the value of 1.93 used in ANL-6302, the BORAX-V Design and Hazards Summary Report.

These plots, and all other flux mapping work, are being analyzed for inclusion in a test report. An effort is also being made to compare calculated with measured flux shapes adjacent to control rods.

After boric acid removal, the boron-stainless steel poison rods were relocated and the number reduced from 36 to 32. The reduction was possible because the more effective new locations are at the flux peaks. The banked control rod position for reactor critical was lowered by about 0.2 inch. The change in available excess reactivity is less than +0.5%. Checks were made to assure that the withdrawal of no single control rod would make the reactor critical. The poison rods will be located as shown in Figure 1 for hot zero power and power operation.

The reactor fuel was removed from the vessel and placed in the storage pit. During unloading, critical experiments were performed with 42, 36, 30, 26, 24, and 22 assemblies in the reactor. With 20 assemblies, the reactor is subcritical with all control rods out. Pertinent critical experiments will be repeated during reloading with the new control rods.

The reactor and systems were prepared for hot preoperational tests by draining and cleaning the boron addition system, rechecking instrument calibrations, and radiographing the new welds made in the plant since the construction contract was completed.

Approval was received to operate both the boiling and superheater cores at powers up to 40 Mw thermal.

Figure 1. Final Poison Rod Location Reference Boiling Core BORAX-V

2. Procurement and Fabrication

a. Superheater Fuel - Inspection of all 230 HPE (half-enriched peripheral) superheater fuel plates has been completed with three rejected and 227 accepted. To date, 14 FPE (fully-enriched peripheral plates) have been inspected and accepted.

Thirty-one 4-plate enriched fuel elements for the central superheater have been vacuum-brazed with Coast Metals 60 alloy. The completeness of the braze joints along the longitudinal square spacer wires has been inspected by leak-testing each coolant channel at 20 psig argon with the element immersed in water. To determine unbonded area, the gas leakage flow rate was calibrated against known flow areas. No leaks were found in 70% of the channels tested; the remainder showed a few minor unbonded areas per element. The largest unbonded area found is estimated to be equivalent to an area 0.0002×0.062 in. It has been calculated that these unbonded, brazed areas will cause only a small local rise in fuel plate temperature beneath the spacer wire; therefore, the fuel elements are considered acceptable. However, procedures are being developed by Central Shops for repairing the leaks which have been detected in 11 of the fuel elements. Production brazing of enriched superheater fuel elements is continuing.

Two 5-element central superheater fuel assemblies have been welded and appear to be satisfactory in all respects.

b. Reactor Components - The poison sections of the new control rods made of 50 w/o Boral canned in Type 304L stainless steel were subjected to a second 4-day period of autoclave testing at 600 psig, 486°F conditions, making a total of 11 days of autoclave testing.

No leaks were detected in any of the 11 sections tested, and the cruciform rods were found to be satisfactory. However, the 5 T-shaped sections were found to have a transverse bend in the 14-in.-wide blade with edges deflected toward the 7-in. blade. The maximum deflection measured was about $\frac{1}{8}$ in. from flat. To determine if this bending was purely a thermal phenomena, the T-shaped rods were heated in a furnace to 500°F and the 14-in. blade deflection measured at high temperature and again at room temperature after cooling. No change in deflection was noted due to temperature changes alone.

The 14-in.-wide blades of the 5 T-rods were given a reverse bend (edges deflected away from the 7-in. blade) of $\frac{3}{32}$ in. and re-autoclaved at 600 psig saturated conditions for 4 days. The wide blade deflection after autoclaving varied from 0 to 0.085 in. reverse bend. Maximum deflection at high temperature and pressure, as measured on one rod by a fixture with friction gauges, showed no movement in excess of the final post-cooling deflections.

To verify dimensional stability, the "T" sections were returned to the autoclave for another 4 days at temperature and pressure. After this test, measurements showed that the wide blades of four of these rods had an average angular movement toward flat of 0.005 in. with a maximum movement of 0.021 in. The other T-rod had a maximum movement of 0.048 in. The four best T-shaped poison sections are considered satisfactory, and are being assembled with followers and extensions.

The new cruciform control rods have been completely assembled with X-8001 aluminum followers and stainless steel cruciform extensions, and inspected.

The dummy control rod with the new design of follower-to-cruciform extension mechanical joint has been scrambled a total of 2,000 times at 600 psig, 486°F conditions with no swelling, bowing, or observable damage of any kind to the joint or control rod. This test has been discontinued.

c. Experimental Components - The radiolytic gas recombiner equipment was moved from the BORAX-V reactor building and installed on the main floor of the EBR-I building. Several modifications and improvements were made, based on previous experience obtained while the equipment was operating in the BORAX-V area. Cold hydrostatic testing of all components except the Knallgas (H_2 - O_2 production) system has been completed.

New instrument racks have been purchased. The electronic instruments servicing the sampling station (exclusive of the fission product monitoring instruments which are being relocated to the heating and ventilating building) have been installed in one rack. The controls for the movable, dual-purpose steam probe and high temperature neutron source drive are also mounted in this rack.

Fabrication of the variable speed oscillator drive was completed. Fabrication of the oscillator rod (Boral canned in Zircaloy-2) continues. The poison section is now being autoclaved. The oscillator must still be assembled and balanced prior to testing in the reactor. The three oscillator guide brackets are complete.

Mechanical stops have been set on the high temperature source drive. The unit was also test run in the vessel at atmospheric conditions.

Material for the step function generator pressure thimble has been procured and fabrication has started. A study is being made to determine the feasibility of using the electron beam welder to join the Zircaloy-2 to the Type 304 stainless steel.

An order has been placed with General Electric Company for a standard in-core miniature fission chamber for use in high power flux mapping and transfer function measurements.

Helium leak tests on the pressure seal NIOIRA braze joints for the sheathed thermocouple leads from the initial instrumented boiling fuel assembly indicated leaks through both seal plates. The seal block was rebrazed and helium leak tested again. The bottom seal plate brazes were satisfactory, but the top seal plate still leaked. In addition, four thermocouple sheaths broke off flush with the top of the upper seal plate. Examination of the sheaths indicated that the sheath walls had necked down and were very thin at the point of failure. Methods of salvaging the thermocouples in this instrumented assembly and the use of thermocouple packing glands instead of a brazed seal are being investigated.

A recorder has been modified for readout of in-core superheated steam and reactor vessel specimen temperatures. Assembly of the wiring harness and reactor pit wire support booms for the instrumented boiling fuel assemblies was completed.

3. Design

Detailing continues on the design of the "head-on" fuel handling tool, and design has been completed for an alternate auxiliary steam line from a 3-in. reactor head nozzle.

Detailed design was completed on the in-core pressure thimble assemblies for flux wires and for miniature fission chambers. These thimbles will be used for high power flux mapping.

The design of the pressure seal-terminal box assembly for the instrumented boiling fuel assembly has been revised to use the thermo-couple packing glands instead of a brazed seal.

4. Development and Testing

a. Plant Tests - Actual cold measurements taken with the boiling core structure and the reactor vessel head in place gave an average deflection on the natural-convection Belleville spring of 0.083 in. and resultant hold-down load of 6,500 lb. Using a calculated differential expansion between reactor vessel and core structure of 0.143 in. when at operating temperature and pressure, gives a hot natural-convection Belleville spring deflection of 0.226 in. with a 20,600 lb hold-down load on the core structure.

b. Corrosion of Structural Materials in Superheated Steam - Samples of stainless steels (304, 310, 316 and 347) have been corrosion tested in 540°C, 600 psi, deoxygenated steam for 105 days. The surface appearances were good, with 304 being perhaps the least resistant. Total weight gains were between 1 and 2 mg/cm² for all samples.

A corrosion product spalling phenomenon in stainless steels exposed to 650°C, 600 psi steam is being investigated. To date it is apparent that such spalling does not occur in two weeks in deoxygenated steam, whereas in oxygenated steam it occurs almost immediately. The steels in this group are 304, 316, and 347 stainless. Certain other minor differences between the testing conditions are presently being eliminated.

A sample of 406 stainless steel also currently in test at 650°C, 600 psi in oxygenated steam had shown good corrosion resistance and little or no product spalling. However, this month a very small area of flaky, white product was noted on the sample, owing possibly to Al in the alloy or to an inclusion. Another 406 sample has been added to the test.

c. BORAX-V Superheater Critical Experiment - The BORAX-V superheater fuel elements are to be used for a brief critical experiment scheduled to start near the end of June. Other components are all on hand. These experiments will be carried out in the ZPR-VII facility. The Hi-C core already loaded in ZPR-VII will be unloaded a week in advance to permit the changeover in order to accommodate the fuel element superheater loading.

II. LIQUID METAL COOLED REACTORS

A. General Research and Development

1. ZPR-III

a. Experimental - Measurements on the spherical Assembly No. 39, the spherical model of a large core with aluminum as the principal diluent, have been completed and the machine unloaded. The shape factor for Assembly No. 31 (a 425-liter cylindrical core with U^{238} , stainless steel, and aluminum diluents) based on a critical mass of 426.5 kg U^{235} for Assembly No. 39 is 0.921, with an estimated error of ± 0.002 . For a reactor of closely similar composition to Assemblies 31 and 39, Loewenstein and Main calculated a shape factor of 0.92 (ANL-6403).

The following measurements were made on Assembly 39 during the period of this report:

(1) Effect of "Step-Wise" Construction of the Core - The spherical shape of the core was approximated by constructing a series of horizontal discs of thickness equal to the 2-in. depth of a matrix cell. Due to the differences in diameter of these cylindrically shaped layers, there are 2-in. vertical steps in the core surface varying in length. The effect of changing the size of these steps at one of the "polar caps" was measured. The radii of the various cylindrical layers of core material are given in Table I. In the upper two layers, the 2-in. steps were replaced by 1-in. steps.

Table I. Radii of the Layers of the Core

Row	Original Radius (in.)	Radius After Smoothing (in.)	
H	4.35	H upper	1.83
		H lower	5.88
I	9.31	I upper	8.30
		I lower	10.16
J	12.17		
K	14.15		
L	15.58		
M	16.61		
N	17.31		
O	17.72		
P	17.85		

The change was made without altering the total mass of fuel involved. The reactivity increase was 9.8 ± 0.5 lh, equivalent to 0.1% of the critical mass.

A subjective estimate of "roughness" indicates that it is about the same for both the cylindrical and spherical core and, in view of the small effect of smoothing a "polar cap" of the spherical core, it is felt that the shape factor of a truly cylindrical version of Assembly 31 has been determined within approximately 0.2% and is $0.921 \pm .002$.

(2) The Worth of Core Material at the Core Edge - The worth of U^{235} , with associated core material, at the edge of the core was found to be 25 lh/kg U^{235} (average value).

(3) Reactivity Coefficients - The reactivity coefficients of sulphur, tungsten, and thorium were measured at the center and near the edge of the core (radius 13.1 in.). Measurements are shown in Tables II and III. In addition, reactivity coefficients for stainless steel, beryllium, polyethylene, enriched uranium, and plutonium were measured at the core center. The results for U^{235} and Pu^{239} agreed with those for Assembly 31.

Table II. Central Reactivity Coefficients of Nonfissile Materials (Assembly 39)

Material	Sample Worth	Reactivity Coefficient
Type 304 Stainless Steel	- 1.50 $\pm$ 0.5 lh	- 1.47 $\pm$ 0.5 lh/kg
Sulphur*	- 1.35 $\pm$ 0.6	- 6.76 $\pm$ 3.0
Tungsten*	-14.35 $\pm$ 0.6	- 7.69 $\pm$ 0.3
Thorium	-14.75 $\pm$ 0.5	- 9.76 $\pm$ 0.3
Beryllium	+22.50 $\pm$ 0.5	+93.5 $\pm$ 2.0
Polyethylene	+17.50 $\pm$ 0.5	+1204.0 $\pm$ 34

*These samples are canned in stainless steel for which the sample worth has been corrected.

Table III. Reactivity Coefficients of Nonfissile Materials at a Radius of 13.1 Inch

Material	Sample Worth	Reactivity Coefficient
Type 304 Stainless Steel	+2.50 $\pm$ 0.5 lh	+2.45 $\pm$ 0.5
Sulphur*	+0.36 $\pm$ 0.6	+1.80 $\pm$ 3.0
Tungsten*	-2.75 $\pm$ 0.6	-1.47 $\pm$ 0.3
Thorium	-3.60 $\pm$ 0.5	-2.38 $\pm$ 0.3

*See footnote to Table II.

(4) Fission Rate Ratios - Fission rate ratios for U^{233} and U^{238} , relative to Pu^{239} , were measured at the core center of Assembly 39 and found to agree within 1.5% with those for Assembly 31.

(5) Radial Fission Rate Distributions - Horizontal radial fission rate scans were made with enriched and depleted uranium fission chambers. One set of traverses was made along a radius parallel to the planes of the fuel plates and another set perpendicular to the fuel planes. The main object of these measurements was to detect any appreciable asymmetry of the flux distribution due to heterogeneity of the core; no such effect was detected.

b. Theoretical

(1) Discrete Ordinate Quadratures for Thin Cells - It is necessary to use a high-order DSN approximation, i.e., many discrete angles, with thin lattice cells, in order to obtain sufficiently accurate flux solutions to the ZPR-III fast critical heterogeneity calculations. The calculations are reported in ANL-6218.*

An analysis of the problem and comparative calculations using both single Gaussian and double Gaussian quadratures are reported in ANL-6345.**

Two different quadrature assignments are being considered for use with the discrete ordinate codes. One quadrature assignment is based on a modified form of the single Gaussian and the other is based on a first collision density angular distribution from integral transport theory. These have been designated MSG, for the modified single Gaussian, and TRIM, for the thin-region integral method. For the case of the thin cell reported in ANL-6345, the use of the MSG and the TRIM with six discrete angles give an error of about -5% in

$$\left[\left(\frac{\bar{\phi}_{\text{source region}}}{\bar{\phi}_{\text{non-source region}}} \right) - 1 \right]$$

By comparison, the double Gaussian method using six angles gives an error of about -26%.

(2) ZPR-III Shape Factors - Shape factor calculations for the ZPR-III assemblies Nos. 23, 32, and 31 carried out using the Los Alamos TDC code were reported in ANL-6565 (Progress Report, April, 1962).

*D. Meneghetti and M. F. Loomis, "Calculation of Heterogeneity Effects in ZPR-III Fast Assemblies Using the DSN Program," ANL-6218 (November, 1960).

**D. Meneghetti, "Convergence of Transport Solutions for Thin Slab Cells," ANL-6345 (April, 1961).

Analogous calculations using diffusion theory have since been undertaken using the one-dimensional RE-122 code for the spherical geometry and the two-dimensional PDQ code for the two-dimensional R-Z geometry. The shape factors calculated by diffusion theory differ by about +0.1%, -0.4%, and +0.4% respectively from the previous TDC-S₄ transport calculations.

2. ZPR-VI and ZPR-IX

a. Reactor Cells - The painting of the exterior walls of the two reactor cells was completed. The leakage tests on Cell No. 4 have been suspended until the damaged rubber door seals on the personnel and emergency doors are replaced. The door seals were damaged during the leakage testing program.

b. Bed and Table Assembly - The installation of the bed and table assembly and the matrix for the ZPR-VI assembly is virtually complete. The final alignment of the matrix tubes will follow the checkout of the wiring of the table drive system.

The assembly of the supports for the ZPR-IX matrix and control rod drives has been initiated.

c. Console and Nuclear Instrumentation - Except for a few minor components, the ZPR-VI facility is wired to the console. The vendor is now inspecting and testing the wiring systems.

The log N, picoammeters, and period meters which did not meet specifications have been returned to the vendor for correction.

The wiring of the ZPR-IX facility is in progress.

All of the coaxial cables for nuclear instrumentation between the console and the cell have been installed. Checking for continuity and noise in the cables is now in progress.

d. Control and Safety Rods - Modifications are being made on the dual-purpose control and safety rod drive mechanisms to improve the performance of the spring action in the scram drives. Rod travel times are being measured from motion picture records.

e. Miscellaneous

(1) Fuel Plates - The enriched uranium fuel plates are being inspected for orientation of rolling. A random sampling of 300 plates or 10% of the $\frac{1}{16} \times 2 \times 3$ in. plates were checked. All of the 300 plates were correctly oriented. Approximately 300 non-randomly selected $\frac{1}{16} \times 2 \times 2$ in. plates

were also inspected. In about 100 of these plates the arrow indicating the direction of rolling was pointed in the wrong direction. Random sampling inspection of the 2 x 2 in. plates is being continued.

The coefficient of expansion was measured on a number of the enriched fuel plates. An average value of 8.3×10^{-6} per degree C was obtained for the direction of rolling and 16×10^{-6} per degree C in the direction perpendicular to the direction of rolling.

(2) Solid State Detectors - Surface barrier solid state detectors coated with fissionable material are being developed for use in making fission ratio measurements. The resolution of these solid state detectors for alpha particles from Cm^{244} has been found to vary from 14 to 25 kev (full width at half maximum) depending on the surface area. The efficiency of the U^{235} -coated detectors to thermal neutrons has been found to be better than 2.5%.

3. AFSR

The defective uranium piece whose pimpling resulted in shutdown last fall was successfully recast. Rejacketing of the two top core sections was completed during the first week of May. These two sections, along with uncanned incremental discs for the lower section, were received in Idaho on May 31 for a new critical mass determination, after which the lower section will be jacketed.

Previously, sample prototype welds of jacketing material were received, inspected, and approved, and a rigorous out-gassing and leak-testing procedure was agreed upon and performed.

It is not believed that this revision will add to the hazards originally analyzed in the Hazards Summary Report ANL-6024 for the following reasons:

1. The core will be clad in 18 mils of 304 stainless steel instead of five mils of nickel. This greatly increases the mechanical and oxidation protection of the fuel. Due to the increased amount of clad material in and around the core, there will be an increase of approximately 1 to 2% in the amount of uranium actually loaded, depending on whether it is necessary to add one or two disc fuel increments.

2. The U-Fe eutectic temperature (725°C) is negligibly lower than that of U-Ni (738°C). In the critical region of the glory hole, there is still a zirconium shell between the uranium and the glory hole liner as described in the hazards report.

3. Elimination of the foil recess substantially improves the canning of the lower section and reduces the likelihood of a highly reactive sample being inadvertently introduced into the reactor.

4. There are no changes in control mechanisms or in safety instrumentation or in operating procedures. Assembly and return to operation will be by means of the same general procedure as outlined and executed when the reactor was first placed in operation; i.e., after loading the upper (stationary) part of the core, the reactor will be brought critical by conventional stepwise addition of fuel in the lower section. When the exact operating load is determined, the individual discs in the lower section will be canned in a single can. Final operating critical condition will then be approached stepwise by means of the shim rods.

Some preliminary tests have been made with the methane-filled neutron telescope by means of a Pu-Be source. The device appears to function as expected and work is continuing in the preparation of equipment to measure fast neutron spectra.

A reactor "criticality meter" has been designed for determining the degree of subcriticality of a reactor. Procurement of equipment for a prototype is under way.

B. EBR-I

1. Mark III Operation

The reactor was operated for approximately half-time during the month at high temperature and high power for the purpose of training personnel and testing a fission-break monitor being developed by the Laboratory.

A cladding failure monitor has been designed, constructed, and successfully tested for use in EBR-I. The monitor operates on the principle that in the event of a cladding failure small quantities of the rare gas fission products, Kr and Xe, are released to the argon gas blanket above the reactor. A supply of the cover gas is passed continuously through an electrostatic precipitator which operates at a potential of 2500 volts. Three major radioactive species are present in the cover gas: A^{41} , and various isotopes of Kr and Xe. Discrimination against the A^{41} , which appears as an intolerably high background, is afforded by the precipitator. At the instant of beta decay, each of the rare gas atoms becomes positively charged and is attracted to the negatively charged center wire. At the time of fixation, the A^{41} has transmuted to stable K^{41} , while the Kr and Xe atoms have transmuted to Rb and Cs atoms, respectively, each of which is still beta unstable. The center wire, which moves at a speed of approximately 18 in. per minute, passes through a mercury gas seal which has

floating on its surface a few cubic centimeters of water. The strongly electropositive Rb and Cs species react instantly with the water and are permanently fixed in the hydroxide form.

In the preliminary experiments, the NaI crystal multi-channel pulse height analyzer was used to analyze the gamma radiations emitted from the water source. The results show that almost perfect discrimination against the A^{41} background was attained and that the net response was particularly sensitive to rare gas species emitted during cladding failure. Additional experiments intended to test the sensitivity of the method and to calibrate the equipment in terms of an actual failure are planned.

2. Fabrication of Core IV Fuel

Zircaloy tubing jackets were received adequate to complete the 420 fuel rods, and 11 of the required 15 Zircaloy thermocouple wells. A total of 62 acceptable fuel rods and 11 acceptable thermocouple fuel rods were produced during the reporting period. This included one special, unbonded thermocouple rod for use in breeding-gain experiments. A total of 497 jackets were loaded, NaK filled, bonded, welded, and inspected, to produce 420 acceptable fuel rods. The 120 blanket rods had been completed previously (see Progress Report, March, 1962, ANL-6544).

After assembly of the eleven Zircaloy-2 thermocouple rods, three rods showed NaK levels somewhat higher than specified but were still acceptable. Compensation for NaK level was somewhat more difficult for the thermocouple rods than for the fuel rods. The fuel rods were overfilled under vibration and the levels were adjusted by drawing off the excess NaK. With the thermocouple rods it was necessary to adjust the NaK level before insertion of the thermocouple well and to compensate for the displacement of NaK by the well.

The resulting sodium level, before insertion of the thermocouple well, was below the top of the drilled upper blanket slug. The thermocouple rod was first overfilled with NaK. Adjustment of the level was accomplished by use of a bent tipped needle with a sliding stop. The desired sodium level, measured from the top of the upper blanket slug, was calculated using the measured dimensions of the slugs and of the thermocouple well.

The needle was inserted to contact the blanket slug, and the stop adjusted so that the needle could be inserted into the hole in the slug to the desired level. The excess NaK was then sucked from the rod. The thermocouple well was slowly inserted and the collar on the thermocouple well was heliarc-welded to the coupling fitting at the top of the jacket. The thermocouple rod was then bonded by heat treating at 300°C and the NaK level was determined by X-ray.

3. Production of Core IV Breeding Gain Specimens

A total of 2560 specimens of natural and enriched uranium, fuel grade plutonium, high isotopic purity Pu^{239} and U^{235} (free of U^{236}) were desired for breeding gain measurements for EBR-I Core IV. In addition, six special fuel rods, 25 insertion thimbles and 51 "baskets," or specimen carriers, were required.

The February Progress Report (ANL-6525) described some techniques proposed for the production run. However, difficulty has been experienced in the capacitor discharge and heliarc welding of the 0.020 in. aluminum jackets on the 0.156 in. diameter x 1 in., plutonium-1.25 w/o, aluminum slugs. Because of inconsistent welding results, recent efforts have been directed toward the development of pressure welding techniques in which the aluminum can material is flowed over the top of an aluminum plug.

The steps required to accomplish this are shown in Figure 2. The resulting assembly is pressed in a coining die to dimension the jacketed slug, to bring the jacket into intimate contact with the core, and to produce a pressure welded seal at the top of the can. Best results are achieved by coining in two stages. A projection was left on the top plug. The top plunger of the coining die was machined with a hole for this projection.

Figure 2. Assembly, Jacketing & Pressure Welding Breeding Gain Specimens

The first upset caused jacket material to flow over the edge of the plug and to be extruded around the projection. The second upset was done with the flat coining plunger which caused the projection to flow out under the pressure forming a rivet-head-type of closure. Preliminary work,

done with unhardened dies, limited the pressure which could be applied to about 50,000 psi. Hardened dies are being fabricated which will allow pressures of 100 to 200 psi to be used to improve the weld consistency.

The dies have been received for punching the 0.125 in. diameter x 0.0043 in. thick, plutonium-1.25 w/o, aluminum foil samples. An experimental shearing, edge welding, and coining die was made to encapsulate the plutonium foils in 4 mil thick aluminum jackets. The first results appear to be successful. Tests are under way to determine the adequacy of the welds produced.

4. EBR-I Mark V Conversion

The detailed drawings required for the construction of the in-pile Mark V loop are in the final stage of completion. A 11.5-kw tungsten filament heater was built and tested for possible use on a prototype out-of-pile test loop. Various ceramics are being investigated for use as electrical insulators in advanced furnace design.

C. EBR-II

1. Reactor Plant

Major activity in the reactor plant for the month centered around completion of miscellaneous jobs which were interrupted or postponed by the Dry Critical Experiments, and a few minor modifications for which a need became apparent as a result of further testing or study. The following are typical of these jobs:

a. Shutdown Cooling Systems - These two systems have been completely helium leak tested, filled with NaK, and are ready for operation.

b. Helium Leak Testing - All nozzles of the primary tank cover have been successfully helium leak tested and the shutdown cooler systems filled with NaK. A reactor vessel cover locking mechanism nozzle will be tested after the completion of mechanical work on the vessel.

c. Temporary Cold Trap System - A temporary cold trap system has been installed in the reactor plant. This system is necessary to augment the permanent sodium purification system during initial cleanup of the primary sodium subsequent to filling the primary tank.

d. Primary Tank Heaters - The combined capacity of all six primary tank nozzle heaters is 670 kw as originally installed. However, installed feeder capacity for the heaters was only 320 kw. Modifications are now being made to add an additional 320 kw of feeder capacity.

- e. Flexible Seals - Installation of the flexible seal assemblies required for the secondary system piping at points of penetration into the reactor building shell is in progress.
- f. Argon System - A final operational checkout of the argon system has been completed and the system is now ready for purging.
- g. Emergency Air Compressor - A compressor unit has been installed in the reactor plant to supply emergency instrument air to the reactor plant in the event of loss of the plant instrument air supply or closure of the isolation valve on the instrument air inlet pipe. Power for the compressor can be supplied by a 100-kw diesel generator set or by the 400-kw emergency diesel generator set.
- h. Nuclear Instruments - Installation of nuclear instruments in the "O" and "J" thimbles has been completed.
- i. Safety Rod Drive - New case hardened gears have been installed on the power take-off shaft of the safety rod drive. These replaced the original carbon steel gears which were showing signs of wear as a result of operation of the safety rod drive. In addition, the design has been modified to permit manual operation of the drive to facilitate changing the safety rod guide tubes. Originally, while replacing the guide tubes, it was possible to scram the safety rod drive while the safety rod was engaged by the fuel handling gripper mechanism. The modification permits the drive mechanism to be disconnected from the drive shafts prior to disengaging the safety rods.
- j. Fuel Element Rupture Detection System - The collector and piping for the fuel element rupture detection system which is in the bulk sodium of the primary tank has been completely installed.
- k. Replacement of 17-4 PH Steel - One part of each of the twelve control rod drive grippers was initially fabricated of 17-4 PH steel. This part, the jaw actuator shaft connector, has now been replaced in all twelve grippers with a similar part made of ASTM Type 304 stainless steel.

In the safety rod drive mechanism, the original 17-4 PH steel power take-off shaft has been replaced with one made of SAE-4140 steel.

- l. Holddown Guide Segments - Holddown guide segments were installed along the periphery of the top of the center reactor blanket. The tops of these devices are identical to the tops of the subassemblies and serve to provide circumferential support to the holddown mechanisms when moving subassemblies located in the outer row of the outer blanket.

m. Thermal Baffles - Additional thermal baffles have been installed around the inner liner of the reactor vessel to reduce potential thermal stresses resulting from reactor scram.

n. Fuel Unloading Machine Subassembly Gripper - Minor modifications have been made to the fuel unloading machine subassembly gripper as a result of recent tests in sodium. Following these modifications, testing of the gripper operation in sodium was resumed.

o. Reactor Vessel Cover Lock Mechanisms - New lower bearings were installed in the reactor vessel cover lock mechanisms. These are aluminum bronze bearings, replacing others which had caused some galling of the shafts. A new aluminum bronze lower bearing for the transfer arm was also installed to replace the original bearing which had caused galling of the transfer arm shaft.

p. Storage Basket - Checkout of the storage basket indicated excessive movement of the drive superstructure with large (0-60 subassemblies) variations in the basket loading. To eliminate this undesirable motion, additional superstructure was added to stiffen the assembly. A counterweight was also added to aid in lifting the basket load and reduce the movement of the superstructure.

q. Cleanup of Primary Tank - Final cleanup of the primary tank and all its interior equipment was started.

2. Instrumentation

a. Fuel Element Failure Detector for EBR-II - A facility which duplicates conditions under which delayed neutron emitters present in fission products in the coolant of EBR-II can be detected was described in ANL-6473 (Progress Report, November 1961). A sodium source of about 10 curies has been activated in CP-5. The source, the D₂O-filled tubes which provide neutrons by the (γ ,n) reaction, the graphite stack, and associated detection and electronic equipment constitute the basic elements of the test facility.

After preliminary tests, measurements of the integral and differential spectrum of a number of different thermal neutron detectors were made, both for neutron plus gamma background and for pure gamma background. The specific radiation is controlled by raising the source in the central tube, and raising or lowering the D₂O neutron-generating annulus for neutron plus gamma and gamma runs, respectively. The neutron flux, thermal and epithermal, was measured with indium foils with and without cadmium covers at 12 points inside the graphite stack, and the gamma source was measured absolutely by coincidences. Fast as well as slow

electronics were tested, although some of this work must be repeated because of equipment malfunction during the experiment. The data should permit calculation of conditions at the fuel element failure detection station within an uncertainty factor of 20%.

In the near future, the response of other detectors, especially some miniature BF_3 chambers and the first model of the special fission chamber which has been developed for this project, will be determined. The transistorized electronics still require a final checkout and the effect on the signal of varying size and geometry of the graphite stack may be investigated.

3. Power Plant

Non-return valves were installed in the steam lines to feedwater heaters Nos. 3 and 4.

Modifications to the level alarm connection on feedwater heaters Nos. 3 and 4 were completed.

Eighteen hundred gallons of new turbine oil was furnished by the supplier to replace the original oil. It was placed in the turbine after draining and carefully cleaning the main turbine oil tank, seal and bearing drain enlargement, seal oil storage tank, and the motor-driven feedwater pump oil tank.

Changing and correcting the pipe hangers in the Power Plant is almost complete.

4. Sodium Boiler Plant

Insulating and installing ferritic steel jacketing on stainless steel lines, induction heating wire, sandblasting exposed ferrous surfaces, and coating and painting were continued. The final tie of one portion of the steam, feedwater, and blowdown pipes to the power plant piping was completed. Electricians completed their work on the control and control panel modifications and the ADT system. Hydrostatic test of the steam, feedwater, and blowdown systems was accomplished.

5. Fuel Cycle Facility

a. Construction and Installation - Corrective work is continuing in the EBR-II Fuel Cycle Facility. The sealing surfaces of the large and two small transfer locks between the Air and Argon Cells are being repaired to eliminate leaks. Corrective work on the Argon Cell cranes and manipulators, made necessary because of the contamination of the bearings with dust, is being continued. The work has progressed to the point where one of the two cranes and one of the six manipulators are now in use.

The void under the Air Cell floor (see Progress Report, April 1962, ANL-6565, page 24) has been filled by pressure grouting. A 24-inch diameter by 10-foot long pipe has been installed in the floor of the Air Cell. This pipe will be used to observe whether any further settling of the fill will occur.

Work was started on installation of the decanner valve bank assembly, and wiring of relay cabinets in basement cubicles for control of refabrication equipment.

Work continued on high-frequency power distribution, compressor discharge header, furnace off-gas installation, pressure-vacuum system, large transfer lock, transfer cell equipment, installation of "feed-throughs," and installation of dry argon compressor.

Instruction of five staff engineers and three technicians in in-cell crane and manipulator operation has been completed. This permits the operating group to install cell lights, removable window sections, service sleeves, and process equipment by remote techniques. These operations are now in progress under conditions effectively simulating those of hot operation for the purpose of defining problems and needed modifications. Crane and manipulator training of additional technicians has started.

Tests are being conducted with the melt refining furnace to determine if a 50-50 solder may be substituted for a bismuth-tin alloy to allow a 40-degree higher melting temperature for the freeze seal, inasmuch as the cell ambient temperature will be higher than that of the mockup used for development. Thus far, a good vacuum has been achieved with the solder seal, and heating rates and temperature have been adequate.

Shielding doors for the analytical hot cells in the laboratory and service building are provided with steel jack screws driving SAE 40 bronze nuts for positioning the doors. One of the nuts failed recently by shearing the square threads. Examination of the failed nut, and of another which was removed earlier, indicates that the strength of the bronze was inadequate, although misalignment and rough thread surfaces may have been contributing factors. It is planned to replace the nuts on one of the doors with similarly designed nuts of a higher strength and to run an accelerated service test. Nuts on the remaining doors will be removed for examination.

Shielded coffins are necessary for transferring fuel assemblies between the reactor and the Air Cell of the Fuel Cycle Facility. Both Argonne National Laboratory and a commercial manufacturer are designing coffins for this purpose. The Argonne design is based upon transferring heat from a circulating argon coolant to the massive lead shielding of the

coffin, whereas the commercial design depends on the use of an argon-to-air heat exchanger. It is planned to have both types of coffins fabricated.

One of the main problems in the use of remotely controlled manipulators in a large hot cell is the perception of depth. While most operations in the EBR-II fuel reprocessing cycle will be conducted within about five feet of a shielding window, some cell maintenance operations will be accomplished by means of a manipulator at distances up to 18 feet from the window at which the operator of the manipulator is stationed. To furnish magnification and to enhance depth perception, a stereo-optical depth perceptor has been designed and four such units will be fabricated.

Locating pins are necessary for the remote installation of replacement components such as the crane bridge drive units. A new locating pin has been designed which overcomes problems of binding that were encountered with previous pins. The new locating pin design consists of a long and a short pin. The long pin engages two shallow holes placed in line before the short pin engages a single hole. This locating pin design has been successfully tested.

b. Development of Remote Control Methods and Equipment for Fuel Fabrication - Design work was completed, and purchase contracts obtained on the settling machine, fuel rod closure welder, bonding machine, and remote control balance. Most of this equipment is now under construction by the vendors. Design work was completed, specifications prepared, and procurement action was begun on the fuel rod closure welder, bond and level inspection machine, and specialized remote control tools. Drawings were completed on the fuel element construction, welding and testing equipment. The final check of these drawings is under way and specifications are being prepared.

The injection casting furnaces have been tested out, desirable modifications have been made, and three sets of bronze, mock fuel pins have been cast. Job descriptions for the installation of the injection casting furnaces have been written.

Work on the fuel pin process machines was inspected on May 22. Progress was disappointing. The vendor had originally promised May delivery. Many parts remained to be machined, subassembly work was just beginning.

6. Training

The Technician Training Program resumed on May 2. Supervisory staff personnel conducted one-hour daily sessions on reactor control, fuel handling, and power plant systems. A reactor technology course began on May 9 to provide technicians with a general nuclear background.

7. Fuel Development

a. Fast Reactor Fuel Jacket Development - Efforts to procure refractory alloys in various forms for advanced EBR-II core jacketing materials is continuing. The results are often discouraging. In some cases after long delays, the vendor will request an order cancellation largely because of a lack of a suitable process to produce the desired end product.

The order for the Mo-0.5 w/o Ti alloy tubing mentioned in the April Progress Report (ANL-6565) has been cancelled at the request of the vendor. Only two feet of the 30 feet of tubing ordered had been received. The delivery of 425 feet of Nb-1 w/o Zr alloy tubing, 0.156 in. ID x 0.009 in. wall x 18 in. long, was made during the month.

The 150 ton vertical hydraulic press and container shift was readied for operation during the month and several extrusion operations were completed including an unsuccessful attempt to extrude a pure Nb billet. The 1 in. diameter x $2\frac{1}{2}$ in. long Nb billet was heated to 1090°C. An extrusion ratio of 21:1 was planned but 110 tons of load failed to produce any 0.210 in. diameter rod. Some damage occurred to the punch, dummy block and container which has since been rectified. The failure may have been due to a decrease in temperature of the billet prior to application of load. Further attempts will be made with improvements in hot billet handling techniques.

b. Properties of Uranium-Plutonium-Fissium Alloys- The thermal conductivity of the U-20 w/o Pu-10 w/o Fs (high Zr) was measured. The values ranged from 0.044 cal/cm-sec-°C at 100°C to 0.086 cal/cm-sec-°C at 800°C. A comparative apparatus with an Armco iron standard was used. Forty points were calculated between 225°C and 760°C. The values listed (Table IV) were taken from a curve drawn through the data points and extrapolated to 100°C and 800°C. For comparison purposes, thermal conductivities of U-10 w/o Fs and U-20 w/o Pu-10 w/o Fs (low Zr) are also listed in Table IV.

Table IV. Thermal Conductivity of U-20 w/o Pu-10 w/o Fs (high Zr),*
U-10 w/o Fs, and U-20 w/o Pu-10 w/o Fs (low Zr)*

Temp (°C)	U-20 w/o Pu-10 w/o Fs (high Zr)* k, cal/sec- cm-°C	U-10 w/o Fs k, cal/sec-cm-°C	U-20 w/o Pu-10 w/o Fs (low Zr)* k, cal/sec- cm-°C
100	0.044**	0.038	0.026
200	0.047	0.044	0.033
300	0.051	0.050	0.039
400	0.056	0.056	0.046
500	0.062	0.062	0.052
600	0.069	0.068	0.058
700	0.077	0.076	
800	0.086**		

*high Zr - Fissium with total Zr content, 2.8 w/o

low Zr - Fissium with total Zr content, 0.5 w/o

**extrapolated curve values.

8. Process Development

a. Melt Refining Process Technology - The results of two-small scale experiments to determine the behavior of condensable fission products during melt refining indicate that cesium and iodine account for nearly all the condensable gamma emitters volatilized. In the first experiment, about 90 percent of the iodine and 70 percent of the cesium collected were in a zone between 450° and 700°C on a nickel condenser. In the second experiment, a Fiberfrax bed was used to simulate a Fiberfrax fume trap in the plant-scale equipment. Nearly all the iodine and cesium accounted for were collected on the Fiberfrax at 450° to 1000°C. A small amount of iodine (~0.25 percent) remaining in the skull material after oxidation is expected to enter the skull reclamation process. Barium and strontium remained largely (96 percent) in the lime-stabilized zirconia crucible. Nearly all (94 percent) of the cerium was retained by the skull. Negligible volatilization of tellurium, zirconium, ruthenium, and uranium occurred.

The nitridation rates of irradiated uranium-fissium fuel pins have been found to be affected by the prior exposure of the pins to air. Pins irradiated to about 1.2 percent burnup were completely unreactive to nitrogen at 300°C after about eight days of exposure to air. In experiments with pins unexposed to air, the nitridation rate of a sodium-coated irradiated fuel pin in nitrogen at 500°C was the same as that of an unirradiated, sodium-coated pin under similar conditions. An irradiated fissium pin in nitrogen at 650°C underwent swelling accompanied by rapid nitridation. Subsequent experiments, in which irradiated fissium pins were heated in flowing nitrogen-argon atmospheres, showed self-heating, swelling, and rapid nitridation, starting at about 660°C in 5 and 100 percent nitrogen. Decreasing the nitrogen concentration to one percent mitigated the self-heating and nitridation effects to some extent. At 0.5 percent nitrogen the effects were definitely reduced.

b. Skull Reclamation Process - The first demonstration run of the skull reclamation process has been completed in an inert atmosphere glove box. A charge of 190 grams of oxidized skull was used in the run. Although detailed analyses are not available, an overall uranium recovery of about 90 percent was realized. This recovery is considered good since much of the remaining 10 percent can be recovered from vessel heels.

A beryllia crucible assembly (pressed-and-sintered beryllia crucible in a stainless steel secondary, with the annular space packed with beryllia powder) has shown no significant deterioration after five runs, each duplicating the sequence of precipitation steps in the skull reclamation process (see Progress Report, January 1962, ANL-6509, page 24). The uranium product in two runs was carried through the retorting step. In the near future, beryllia crucibles (1) prepared by a thixotropic casting procedure and (2) fabricated from pressed-and-sintered bricks or tiles will also be tested.

c. Blanket Processing - The separation of the plutonium-bearing zinc-50 weight percent magnesium supernatant from the uranium which has been precipitated by magnesium addition and cooling to 400°C is an important step of the blanket process. The tenth such separation has been completed. Supernatant removals of about 96 percent were obtained in conical-bottom crucibles and removals of 92 percent were obtained in flat-bottom crucibles. To avoid the necessity of washing the uranium precipitate, a minimum supernatant removal of 95 percent is required. Equipment has been obtained for measuring the removal of the supernatant solution on a 5-kg uranium scale.

d. Reduction of Thorium Dioxide - Further experiments on the direct reduction of thorium dioxide to thorium metal by zinc-magnesium alloy in the presence of a suitable flux have shown that the use of tantalum equipment and an argon atmosphere is desirable to obtain reproducible results. Using fluxes containing at least 50 mole percent magnesium chloride, 7 to 15 percent calcium fluoride, and the balance calcium chloride, complete reduction of thorium dioxide was achieved consistently in three to four hours at 750°C.

e. Materials and Equipment - To test properly the corrosion resistance of tungsten and tungsten-molybdenum alloys under skull reclamation conditions required longer exposure times than have been available in tests conducted in tantalum capsules. To avoid the necessity of an enclosed capsule, specimens of tungsten, molybdenum-30 percent tungsten alloy, and tungsten-0.8 percent iron-0.4 percent nickel alloy were fastened to a shaft to form an agitator which was then operated under noble metal leaching conditions (in zinc plus a halide salt flux at 800°C) for 500 hours. Preliminary results indicate little, if any, effect on tungsten and molybdenum-tungsten alloys, but show that serious degradation of the tungsten-iron-nickel alloy occurred.

In a test conducted at 750°C for 500 hours, the addition of 0.8 percent activated charcoal increased the rate of attack of cadmium on 304 and 405 stainless steels, probably because of carbiding.

f. Special Project - The diffusivity of uranium in zinc was found to range from 1.5×10^{-5} sq cm/sec at 625°C to 2.7×10^{-5} sq cm/sec at 800°C, with estimated accuracies of ± 15 percent.

D. FARET

The general engineering and physics parameters for an experimental facility to test the characteristics of advanced fast reactors are being examined in detail.

1. Flow Distribution for Special High Performance Fuel Assemblies

A double inlet type plenum design for the FARET reactor was described in ANL-6565 (Progress Report, April, 1962). The two plenums, one over the other, are connected by means of ports located along the length of the hollow reflector assembly adaptor which passes through both plenums. The distribution of coolant to the inner and outer assemblies may be varied by changing the total area of inlet ports in the reflector adaptors. A maximum of 12 ports, 0.321 in. in diameter, for a total port area of 0.966 in. sq per reflector adaptor may be used.

Calculations to determine the total area of the inlet ports in the reflector adaptors have been made at the peak inlet pressure of 140 psi, a maximum system flow rate of 6000 gpm, and for a core consisting of 52 subassemblies in which as many as one to seven high performance subassemblies may be used. The results of these calculations are shown in Table V.

Table V. Number and Port Size of Reflector Assembly Adaptors
for High Flow Rates in Special Fuel Assemblies

No. High Performance Assemblies	1	2	3	4	5	6	7
Required Total Port Area (in. ²)	16.61	15.65	14.68	13.72	12.75	11.78	10.82
No. Reflector Adaptors	17	16	15	14	13	12	11

III. GENERAL REACTOR TECHNOLOGY

A. Applied Reactor Physics

1. Fast Neutron Spectrum Measurements

One lithium-sensitized solid state neutron spectrometer and its associated equipment has been checked for proper operation and was sent to Idaho for tests. The pulse height distributions observed in the tests on CP-5 have been reproduced in the thermal column of EBR-I.

Preliminary data have also been obtained from measurements made with the solid state neutron spectrometer in a ZPR-III core. A comparison of the raw data with and without correction for background has been made. The comparison indicates the need for detectors having higher neutron sensitivity and for a duplicate detector assembly that does not contain neutron-sensitive material, so that good background measurements may be made concurrently. Consequently, a duplicate of the first detector assembly has been constructed and, upon the receipt of the second set of nuclear diodes, two detector assemblies will be available for making both separate spectrum and background measurements concurrently.

2. High Conversion Critical Experiment

Experimental measurements are being made using the two-zone cylindrical core described in the last Progress Report (April, 1962, ANL-6565, p. 33). Radial and axial flux traverses were made in the Hi-C cylindrical loading (1:3 H₂O:UO₂), using gold, indium, dysprosium, and lutetium. In addition, copper and manganese were irradiated near the center of the core. The detectors were exposed both bare (aluminum) and cadmium covered. The information will be used to determine the neutron energy spectrum and magnitude of the reflector savings.

Data also have been obtained on the fast fission factor in the cylindrical zone. Flux traverses (bare and cadmium covered) were also made in the cylindrical loading of the core, with 3 wt-% enriched and 0.21% depleted uranium foils to evaluate the conversion ratio. Fuel and void worths are also being measured.

The temperature coefficient was measured in the reactor vessel. The propeller type mixer previously used required more space than is available between the reactor vessel and the larger Hi-C cores so it was necessary to use the spare fill pump which is located outside of the tank to provide mixing. The new arrangement places only the return line nozzle inside the reactor vessel. The pump draws 10 liters/sec from the bottom of the vessel and returns it near the midplane of the core.

Treatment of the temperature coefficient data depends on knowledge of the mechanisms which produce a slow loss in reactivity at temperatures higher than room temperature. If caused by progressive bubble formation (perhaps H_2 from corrosion of the 10^6 cm^2 aluminum surface exposed), the slow loss should not be included with temperature effects. On the other hand this may be caused by slow temperature equilization in the tightly packed core. The drift was not affected greatly by stirring of the moderator or by dichromate addition to reduce corrosion.

3. Theoretical Physics

a. Mathematical Numerical Methods Analysis - This laboratory is collaborating in testing a translator for the international algorithmic language Algol 60, which has been written for the LGP-30 computer by the Dartmouth College Computer Center. This translator, although not entirely complete, is extremely powerful, and will accept the full generality of Algol 60, except for recursive calls of procedures, (i.e., a subprogram which utilizes itself), calls of arrays by value, own arrays, and variable bounds on arrays. Because of the extreme generality, the resulting programs are somewhat slow for extensive production calculations, but the flexibility of Algol 60, the large number of algorithms which have been published in it, and the excellent checkout facilities provided by the Dartmouth translator make the system an extremely valuable one for developing new methods.

In the course of checking out the translator, three published algorithms (for Chebyshev approximation, for interpolation by continued fractions, and for computing the Kelvin functions ber and bei), and an unpublished algorithm for reversion of power series were translated and run successfully. Certifications of these algorithms have been submitted for publication.

b. COST and CYCLE Codes - The IBM-704 FORTRAN-II program, COST, which is designed primarily to perform an economic analysis based on the converged output data of the related CYCLE code, has been partially revised to allow for more flexibility in the assignment of basic costs.

The CYCLE production series has been completed and all of the extensive results of the CYCLE studies have been employed with this new version of COST in order to provide an economic evaluation of these many systems. This large amount of data is now undergoing evaluation. A portion of the results obtained by use of these codes will be presented at the June meeting of the American Nuclear Society in Boston.

The programming of CYCLE to permit a study of the variation over the equilibrium cycle has been completed. This simplified portion of the program may also be used to study one step burning of systems of

arbitrary initial composition. For example, a thermal reactor using fast reactor blanket elements as fuel and not undergoing recycle operation could be examined using this portion of the CYCLE code.

c. Doppler Coefficient Studies - In a previous Progress Report (February, 1962, ANL-6525) concern was expressed about the possible error in fast reactor Doppler coefficient measurements due to heterogeneity in the experiments. In particular, it was felt possible that the use of large fuel rods separated by sodium regions thick compared to those in the reactor would lead to erroneous results. More detailed calculations have now been made which indicate that these effects are probably small, so that one has more flexibility in the design of experiments than was previously supposed. The calculations indicate that by far the most important effect of sodium is simply to furnish more low energy flux, increasing the Doppler effect in proportion. The flux depression in the vicinity of the 3-kev resonance in sodium results in an increase in the Doppler effect in plutonium relative to that in U^{238} since the plutonium effect occurs predominantly at energies below this resonance. The calculations indicate that the positive Pu^{239} effect expressed as a percentage of the negative U^{238} effect typically increases from about 15% to about 20%. The sodium resonance effect is thus relatively unimportant if the calculations are reasonably accurate with respect to the relative contributions of Pu^{239} and U^{238} to the Doppler coefficient.

B. Reactor Fuels Development

1. Corrosion Studies

a. Aluminum Powder Products - Corrosion tests on aluminum powder products are continuing in an effort to find materials which combine strength and acceptable corrosion resistance. Recently, experimental rod extrusions made by Armour Research Foundation completed a 270 day corrosion test in refreshed distilled water at 290°C. Of the 15 extrusions tested, ten were in excellent condition after that exposure. Corrosion penetration varied between 0.002 and 0.003 in. and no accelerated attack was in evidence. Extrusions in this group were based on either Al-1 w/o Ni-0.5 w/o Fe-0.1 w/o Ti or Al-1 w/o Ni-0.1 w/o Ti powder. The powder had been milled with additives such as 3-8 w/o of $AlPO_4$, SiO_2 , ZrC and SiC.

The five extrusions that failed prior to 270 days were those containing various percentages of Al_2O_3 , a 10 w/o ZrC mix, and a lower temperature extrusion.

b. Zirconium Alloys for Superheated Steam - Eleven selected samples of a series of zirconium alloys, corrosion tested last year, have been continued in test in 540°C, 600 psi, deoxygenated steam since January,

and were recently removed from test for inspection after more than 200 days exposure. The alloys contain nickel, iron, and/or copper in amounts ranging from 0.5 to 3%, and the sample stocks had been prepared according to various rolling and heat-treating schedules. From weight gains and surface appearance it is apparent that stock preparation affects the corrosion resistance of these materials significantly, but no one treatment that is more generally beneficial than another is obvious from preliminary inspection of the results. The rate of weight gain of several samples was accelerated after 120 days, but this did not occur for the hot-rolled Zr-3 w/o Ni-0.5 w/o Fe material. Some edge cracking of the corrosion film was seen microscopically on all samples.

Markedly lower pickup of corrosion product hydrogen observed in Zr-3 w/o Ni-0.5 w/o Fe and Zr-1 w/o Cu-1 w/o Fe in superheated steam at 650°C as compared with 540°C has been reported previously. Electron microscope and electron diffraction studies are being made of corrosion films formed in these conditions. No marked differences are apparent in the electron microphotos. However, small crystals are present, and a number of diffraction patterns of these have been produced. Hopefully, analysis of the patterns will reveal differences which will correlate with the difference in hydrogen pickup at the two temperatures.

2. Ceramic Fuels

a. Uranium-Thorium Sulfide - Through continual change in procedure, the total time required to produce a 250 gram batch of US low temperature mixture has been reduced from 16 hours to about 8. The most significant change has been the lowering of the hydride dissociation and H_2S reaction temperatures from 550 to 400°C. This eliminates any tendency of the fine uranium powder to sinter, resulting in a finer product with a much more homogeneous mixture of the US_2 and alpha-uranium constituents.

Work is now in progress to optimize similarly the production of ThS while building up a small reserve of material. The existence of two hydrides, $ThH_{3.75}$ and ThH_2 , complicates this process somewhat.

Two thermal conductivity samples have been submitted for measurement. The first, a 2 cm diameter by 12 cm tube, was filled with homogenized powder packed to 50% theoretical density and run to 1000°C in a longitudinal heat flow apparatus. The results are now being evaluated. The second sample, a 1 in. diameter by $\frac{1}{4}$ in. thick sintered disk of 90.2% theoretical density, has been sent to Dr. McElroy of ORNL for measurement up to 800°C.

b. Preparation of Plutonium Carbide (See Progress Report, March, 1962, ANL-6544, p. 38) - Two operations were demonstrated during the reporting period. Melts were made to recover finely ground particles. The powders were melted in a tantalum crucible. No difficulties were encountered in this remelting operation. A kilogram production melt of plutonium-carbide was prepared to demonstrate scale-up feasibility. The preliminary results show no indication of scale-up problems. The product of the one-kilogram melt is being analyzed for uniformity and microstructure. These results are not yet in.

3. Fabrication Development

a. Fabrication of Zirconium Alloy Tubing - Zirconium alloy tubing is a desirable structural and fuel jacketing material for use in many types of reactors. However, a common type of defect in this tubing has been the unexplained appearance of small cracks, particularly on the ID, extending for considerable distance inward, resulting from some fabrication malpractice. The objective of this program is to develop techniques for fabrication of defect free tubing in size ranges not commercially available. Work to date has been limited to development of techniques for fabrication of tubing smaller than $\frac{1}{2}$ in. OD.

Efforts are under way to improve "core drawing" procedures for fabrication of Zircaloy-2 instrument tubing. Three machined tubes, 0.375 in. OD x 0.068 in. wall x 6 in. long, were "core drawn" without copper jacket or lubricants to 0.280 in. OD through six aluminum bronze dies. The as-drawn tube surfaces were smooth and free from longitudinal striations. Previous tests had produced no means of preventing bare Zircaloy surfaces from seizing in drawing dies and thus resulting in badly galled surfaces.

A hollow Zircaloy-2 billet jacketed in copper was successfully extruded in the form of a ribbed tube. The extrusion temperature was 850°C and the ratio was 28:1. The tube is in process of evaluation.

b. Preparation of Plutonium Foils - In the Progress Report for March, 1962, ANL-6544, certain development work on Pu foils sandwiched in Al was described. These specimens are to be used as cyclotron targets. The foils were to be approximately $3\frac{1}{2}$ in. x $\frac{1}{4}$ in. x 0.004 in. They were to be bonded for thermal transfer to 6 in. x $1\frac{1}{2}$ in. x $\frac{1}{4}$ in. water-cooled target holders, and covered by a bonded-inplace aluminum foil, 0.002 in. to 0.0025 in. in thickness.

Assembly of these specimens is illustrated in Figure 3. A cast slab $\frac{1}{8}$ in. thick, of plutonium-1.25 w/o aluminum alloy was scalped to 0.090 in. thickness and rolled to 0.016 in. After annealing, pieces $2\frac{1}{2}$ in. x $\frac{5}{16}$ in. wide were sheared from the rolled strip and the surfaces were again scalped to produce pieces measuring $\frac{5}{16}$ in. x $2\frac{1}{2}$ in. x 0.012 in.

Figure 3. Plutonium 1.25 w/o Aluminum Cyclotron Target Specimen

The plutonium-aluminum foils were placed in cavities milled in $1\frac{1}{4}$ in. x 1 in. x $2\frac{3}{4}$ in. aluminum backing plates, and were covered by 0.018 in. thick aluminum cover plates. The contacting areas between the aluminum plates and the cover plates only were coated with powdered silicon. The assembly was placed between dies and bonded at 600°C and 1400 psi for 45 seconds. A restraining picture frame was used on the die to control flow of the metal.

The assemblies were then heat-treated and reduced 60 percent in thickness by rolling at 300°C. The roll-bonded compacts were X-rayed and the core thickness was calculated by dividing the original volume by the projected area of the core after rolling. After marking the core location, the clad specimens were sheared to $\frac{3}{4}$ in. x $1\frac{1}{2}$ in. and the edges were beveled at 45°. The surfaces were carefully cleaned and mill-etched.

A cavity was prepared in each of the target holder plates. This cavity was slightly shallower than the thickness of the corresponding clad specimen. The edges were beveled at 45° to match those of the specimen. The cavity was coated with silicon powder, the specimen was inserted, and the assembly was pressure bonded at 1400 psi, between dies heated at 600°C.

The resulting braze appeared satisfactory. The cladding thickness was measured by using a double-pulse, eddy-current technique. The results were compared with similarly prepared specimens which were sectioned. The bonded areas of the sectioned specimens were judged satisfactory.

The resulting target specimens showed an initial surface contamination of 100,000 and 40,000 dpm respectively. Surface "wipes" were counted at approximately 500 dpm. After etching and ultrasonic cleaning, the levels of surface contamination were reduced to 50,000 dpm and 25,000 dpm with no counts showing on the wipes. Further efforts for decontamination were discontinued because of the thinness of the surface coating. The specimens were inspected and judged to be acceptable.

4. Irradiation Studies

a. Irradiation of Absorbing Materials Dispersed in Glass - Samples containing varying amounts of natural boron carbide dispersed in glass were fabricated for possible irradiation experiments. The experiments will determine the irradiation stability of the B_4C -glass combinations. Three types of glass were chosen for the tests. There were (1) a lead glass which has a softening point of about $400^\circ C$, (2) a soda-potash glass with a softening point of about $500^\circ C$, and (3) a soda-lime glass with a softening point of about $600^\circ C$. The lead glass was chosen specifically for its low softening point. The soda-lime glass has a greater resistance to corrosion than the soda-potash glass, but unlike the soda-potash glass it has a tendency to crystallize if held at higher temperatures for an extended period of time.

The compositions prepared contained from 15 to 90 w/o B_4C . Two weight percent Al_2O_3 , not usually found in these glasses, was added to improve their corrosion resistance. The raw materials used for each glass composition were mixed for two hours in a cone-type blender prior to melting. After melting, each glass was crushed in a steel mortar and ball milled in an alumina vibratory mill. That portion of the glass which passed through a 100 mesh screen was used to make the B_4C dispersions. Compacting tests made on B_4C of various particle sizes indicated that a mixture of 70 w/o of -60 mesh particles and 30 w/o of -325 mesh particles would yield the greatest dry-packed density, approximately 70 percent of theoretical. This mixture of the B_4C plus the glass was mixed in a cone-type blender for one-half hour then blended with alcohol in a porcelain mortar and dried. This procedure produced a coating of glass on each B_4C particle.

The B_4C -glass mixtures were loaded into stainless steel tubes by low frequency vibratory packing. The maximum packing density was 71 percent of theoretical. Type 321 stainless steel was used, since the firing temperatures of the samples fell in the range of carbide precipitation

in stainless steel. The firing temperatures were varied from 550 to 950°C, depending upon the composition of the mixture. All samples were fired in a helium atmosphere and furnace cooled to below their softening point before being removed from the furnace. The samples with a high glass content proved difficult to fabricate. It was observed that at temperatures above 575°C the 15 w/o B₄C-lead glass mixture would expand out of the jacket, while at temperatures below 500°C the glass would not soften completely unless held at this temperature for an extended period of time.

The tendency of the high glass content samples to swell indicated the advisability of high temperature testing of completed samples prior to irradiation. Two samples were chosen for the test. These were (1) a sample containing 85 w/o soda-potash glass with a softening temperature of 500°C, and (2) a sample containing 85 w/o lead glass with a softening temperature of 400°C. The samples were placed in a furnace containing an argon atmosphere and brought to a temperature of 500°C. The samples were held at this temperature for 173 hrs and furnace cooled. This temperature was chosen to allow observation of the effects of prolonged exposure to temperatures both at and above the softening points.

A visual examination of the samples after removal from the furnace failed to indicate any changes. The weight and density changes were so slight as to be considered of no importance. Radiographs of these samples, however, indicated that considerable damage had occurred within the core. The lead glass sample had become somewhat shrunken and distorted in shape, probably resulting from a combination of softening of the glass and trapped gases under pressure. Radiographs of the soda-potash glass sample indicated areas of variable density.

Both samples were sectioned and examined macroscopically. The lead glass sample had a substantial random scattering of voids which varied in size. Some metallic lead had formed at the lower end of the sample. The top area of the soda-potash sample which had a high initial density was relatively unaffected. The area which had a low initial density was very porous with a uniform scattering of voids and a high incidence of cracking.

From these indications the use of a lead glass-B₄C mixture as a control rod material where elevated operating temperatures exist is not promising. Even if the shrinkage problem could be overcome, the precipitation of metallic lead from the oxide in the glass would so change the physical properties of the glass as to render it useless for its intended purpose. The soda-potash glass-B₄C mixture seems promising if the glass in the mixture has a high initial density. Irradiations of the test specimens will be made at temperatures well below the softening points of the glass phases.

5. Nondestructive Testing

a. Ultrasonic Techniques - Imaging Studies - Some imaging characteristics of ultrasonic visualization systems are now under study. Equipment to study these effects using photographic film detection methods is in operation and a cooperative program to develop an electronic pickup tube to detect ultrasound is in its early stages. This latter study, which is aimed at the development of an electronically scanned piezoelectric pickup tube for a television presentation of ultrasonic images, is being made in cooperation with Northwestern University.

Using photographic film methods, it has been possible to visualize ultrasonic field patterns by themselves and after transmission through holes and wire meshes. Because of the high ultrasonic intensity threshold required by film detection method it has not yet been possible to visualize internal flaws within metals. Present efforts on this phase of the program are aimed at determining resolution characteristics using various imaging configurations.

b. Neutron Techniques - Resolution Tests - A second set of resolution data has now been obtained which tends to confirm the results found in the first resolution tests described in the April Progress Report. From the curves of resolution versus screen thickness which have been obtained for most of the direct exposure and transfer converter materials, it will be possible to choose the converter material thickness for optimum resolution results.

The boron and lithium loaded scintillators have not yet been subjected to a detailed study of the resolution versus the thickness. Preliminary results with a B-10, ZnS(Ag) scintillator whose overall thickness was less than 0.001 in. do show a slight resolution gain over the 0.012 in. thick scintillator used in most of our studies. The resolution advantage was gained, however, at the expense of a significant loss in speed. More extensive studies of scintillator thickness versus resolution are planned to see if better resolution characteristics can be obtained while maintaining good speed.

c. Radiographic Techniques - Isotopic Analysis - Some progress has been made in developing methods of isotopic analysis of uranium using a 512 channel analyzer. Several samples of UO_2 were analyzed to determine the enrichment. The UO_2 samples were from a batch of blended material containing depleted UO_2 (0.22%) and highly enriched UO_2 (93%). The average enrichment of the final product was expected to be 10%. Since the oxide has a particle size of approximately 50 microns the final mixture could not be expected to be homogeneous on the atomic level. Therefore samples sent for mass spectrometer analysis gave varied

results between 9% and 11%. This was probably a result of the small sample size used by this method of analysis (of the order of 10^{-4} g). Two 250 mg samples of the blended material (including the binder) were used for the scintillation spectrometer analysis. The gamma radiation from the U^{235} and U^{238} isotopes was used for the determination. The percentage of U^{235} determined on these samples agreed within 0.5% (10.00% and 9.95%). The material was homogeneously blended as shown by the fact that counts of one-gram samples agreed within 0.5%.

C. Reactor Materials Development

1. Radiation Damage in Metals

a. Irradiation of Steel in EBR-I - Two sets of sub-size Izod impact specimens of SA-212B carbon steel were irradiated in EBR-I earlier this year. The specimens were in the reactor for 95.4 Mw-hr of operation, and the specimen temperature was about 140°C. The radial locations of the two sets of samples were 4.3 and 7.6 cm respectively. The calculated ratio of defect production between these two locations is 1.19. The predicted activity of a nickel monitor foil at the inner position is 1.27 times that for the outer position.

The irradiated steel showed an increase in ductile-brittle transition temperature of about 250°F. The difference between the two sets is roughly within the scatter of the data, with the more heavily irradiated steel showing slightly greater shifts. The relationship between the sub-size Izod temperature shift and the NDT shift of Charpy V-notch specimens as a function of irradiation dose has not been established. They are both empirical tests and only by careful control of variables can correlations be determined. It is expected that Charpy specimens can be irradiated in the near future.

b. Irradiation of Copper - Two copper single crystals were irradiated in a manner identical to that used for the two sets of steel specimens. They were pulled on an Instron Testing Machine at room temperature and at liquid nitrogen temperature. Corrections for preferred crystal orientations must be applied before true yield stress values can be obtained; however, the crystals did undergo appreciable hardening.

After the 12.8-hr Cu^{64} had decayed these specimens remained relatively active. Measurements indicated an activity of 25 mr/hr at about 2 in. from the sample and analysis on the 200-channel analyzer crystal spectrometer showed the activity to be Co^{60} . The reaction $Cu^{63}(n,\alpha)Co^{60}$ has a reported cross section of 0.72 mb compared to 0.6 mb for the reaction $Al^{27}(n,\alpha)Na^{24}$, in the EBR-I core spectrum. A rough calculation of

the predicted Co^{60} activity shows this cross section to be of the correct order of magnitude. The ratio of activities between the two copper crystals is 1.28. This compares quite favorably with the nickel activation ratio of 1.27 mentioned above.

This reaction offers interesting possibilities as a neutron threshold flux monitor. Copper is easily obtained and handled, the cross section has a high threshold and is small, but the long half-life of the product makes it adaptable as a monitor for long term irradiations. The thermal neutron cross-sections of Co^{60} are not negligible, but are well known. Additional work with this reaction is anticipated.

c. Dosimetry - The observed foil activity resulting from exposure in EBR-I was compared with calculated values. The ratio of the observed to the predicted values was found to be relatively constant. This implies that errors may exist in the absolute normalization of the $\text{Ni}^{58}(\text{n},\text{p})\text{Co}^{58}$ cross section, in the computed power normalization for the reactor, or in the calibration and interpretation of counting data. Any or all are possible sources of systematic error and additional study will be performed to resolve this factor.

D. Heat Engineering and Fluid Flow

1. Boiling Liquid Metal Experiment

The construction of the forced convection boiling sodium facility is sufficiently complete to permit initial performance testing. The air cooling system and two eddy current level gauges (one in the condenser well and the other in the differential pressure system) are not yet installed. All of the electrical heating and scram circuits have been tested and have operated satisfactorily. The next tests will be heat loss calibrations at temperature, after which the loop will be charged with sodium.

The accurate determination of the liquid vapor interface is fundamentally the most important single piece of information to be obtained from two-phase pressure drop experiments. Therefore, the development of instrumentation to measure these values has been emphasized. The sensitive element of the gauge is a bifilar transformer made by winding 20 B&S chromel wire on a ceramic core and fixing the wire spacings by painting with an aluminum oxide type coating. The coil is operated at 50 kc and delivers an amplified signal of approximately 10 mv/cm of liquid height at room temperature. The element exhibited a small temperature coefficient when tested in a furnace up to 1700°F. In general, the stability and sensitivity characteristics of the sensing element is good but some difficulties have been experienced with the associated electronic components.

E. Chemical Separations

1. Fluidization and Fluoride Volatility Separations Processes

a. Direct Fluorination of Uranium Dioxide Fuel - Studies are being made to optimize process conditions for direct fluorination of uranium dioxide pellets. Another in the series of controlled gas recycle runs was made on an 18-in. deep bed of uranium dioxide pellets in Alundum, the charged bed having a weight ratio, $\text{Al}_2\text{O}_3/\text{UO}_2$, of 0.15. Fluidizing gas at a superficial gas velocity of about 1.3 ft/sec at 500°C was used. Oxygen concentration was held to about 1.3 percent at the reactor inlet and 3.2 percent at the reactor outlet. These gas flow conditions were similar to those used previously to give both good fluorine efficiency and freedom from caking. However, in the present run operational instabilities developed, and the run was terminated to allow inspection of the bed. The bed was partially caked, and there had been some gas channelling. The relatively poor performance of this run has been tentatively attributed to two conditions different from the previous run; namely, a higher rate of formation of uranium hexafluoride and a lower $\text{Al}_2\text{O}_3/\text{UO}_2$ weight ratio. A new series of runs is planned with varying pellet-bed and inert-bed depths.

b. Separation of Uranium from Zirconium Alloy Fuels

(1) Studies of the Hydrochlorination and Fluorination Steps with Down-Flow Fixed-Bed Filters - A fluid-bed volatility scheme for processing enriched uranium-zirconium alloy fuels is being investigated. This scheme involves reacting the uranium-zirconium alloy with hydrogen chloride and subsequently fluorinating the uranium chlorides to volatilize the uranium as the hexafluoride. The effect of hydrogen chloride excess and fluorination temperature on uranium retention by the inert Alundum bed material being used as the heat transfer medium was examined further. Exploratory work with phosgene as an auxiliary chlorinating agent was initiated. The quantity of hydrogen chloride used was 13 and 25 times the stoichiometric requirement in Runs 26 and 27, respectively (see Table VI). Fluorine excesses over stoichiometric amounts were high, about 100. Hydrochlorination and subsequent phosgene treatments were conducted at 400°C. The resulting beds were fluorinated at 400°C and re-fluorinated at 500°C.

Table VI. Effect of Phosgene Treatment on Retention of Uranium by Alundum

Alloy Charge: 240 g Zircaloy Sample (5.1 w/o U)
Inert Bed Charge: 320 g Alundum Type RR

Run No.	Hydrochlorination			Phosgene Treatment			Fluorination			Final Reactor Bed Analysis (w/o)		% Initial Zr Retained in Reactor Bed	% Initial U Retained in Reactor Bed
	Fluid-Bed Temp (C)	Time (hr)	Quantity (X Stoich)	Fluid-Bed Temp (C)	Time (hr)	Total Phosgene Fed (cu ft)	Fluid-Bed Temp (C)	Time (hr)	Quantity (X Stoich)	Zr	U		
26	400	17	13	-	-	-	400	6.5	130	0.13	0.05	0.18	1.3
							500	2	40	0.13	0.05	0.18	0.8
27	400	28	25	-	-	-	400	7	140	0.04	0.04	0.06	1.1
							500	2	40	0.04	0.03	0.06	0.8
28	400	7	5	400	5.2	29.3	400	9	~100	<0.03	<0.03	<0.04	0.8
							500	2	~100	<0.03	<0.01	<0.04	<0.2

Use of the greater excess of hydrogen chloride in Run 27 (25 times the stoichiometric requirement introduced over a 28-hr period) seemed to improve overall uranium removal a very modest amount, 0.04 w/o uranium remaining in the bed as compared to 0.05 w/o remaining in Run 26 in which 13 times the stoichiometric amount of hydrogen chloride was employed. In Run 28 the use of phosgene after a relatively short (about 7 hr) hydrogen chloride treatment with five times the stoichiometric requirement resulted in a lower uranium retention, 0.03 w/o residual. Refluorination at 500°C, after initial fluorination at 400°C, gave improved results: a residual concentration of 0.04 w/o uranium was reduced to 0.03 in the case of hydrochlorinated material, and where the two chlorinating agents were used, the uranium content was reduced from 0.03 to less than 0.01 w/o, the best result achieved to date in a single experiment. The zirconium content (which is believed to be associated with uranium retention) was also reduced where greater amounts of hydrogen chloride were used (typical values, 0.04 w/o residual concentration as compared to 0.13 w/o) and again the lowest value to date was achieved when phosgene was used (<0.03 w/o residual zirconium). The lowest residual uranium and zirconium values correspond to a retention of <0.2 and <0.04 w/o, respectively, of the initial charges of the alloyed elements.

(2) Fluid-Bed Hydrolysis of Zirconium Tetrachloride - Experiments in the six-inch diameter column on the fluid-bed hydrolysis (by steam) of zirconium tetrachloride to zirconium dioxide studied further the effects of lower bed temperatures and higher filter blowback pressure on the operation of the unit. Successive runs were made of 6.0-, 3.5-, and 1.8-hr duration using a zirconium tetrachloride feed rate of 2.2 kg/hr (the proposed pilot plant flowsheet rate). The longest run was conducted at 350°C for the first five hours and at 300°C for the last hour. The last two runs were made at 300°C and the filter blowback pressure was increased from 30 to 80 psig. Excessive pressure rise (buildup across the filters) was experienced at the higher temperature during the first run, whereas smooth operation was achieved at the lower temperature in the last two runs. The higher blowback pressure is also believed to be a contributing factor in the improved operation.

2. Chemical Metallurgical Process Studies

a. Chemistry of Liquid Metals - Measurements of the solubilities of various elements in liquid metal solvents are being continued. The solubility of strontium in liquid zinc was measured. Tentative values were found to vary from about 0.8 w/o at 730°C to 0.001 w/o at 470°C. Tentative values for the solubility of barium in liquid zinc were found to vary from about 2.2 w/o at 800°C to 0.013 w/o at 580°C.

Metallographic analyses of three uranium-zinc alloys prepared by annealing for 53 days at 650°C (see Progress Report, March 1962, ANL-6544, p. 47) indicate that the uranium-rich boundary of the epsilon phase at this temperature is approximately $\text{UZn}_{9.4}$.

Preliminary data on the technetium-zinc system indicate that the solubility of technetium in zinc is probably low. Evidence of a possible technetium-zinc intermetallic phase was found.

In a continuation of the study on the calcium-zinc system (see Progress Report, April 1962, ANL-6565, p. 46) by means of the recording effusion balance, the following intermediate phases were found CaZn_{13} , $\text{CaZn}_{11.0-12.3}$, CaZn_5 , and CaZn_2 . The possibility of an additional phase in the vicinity of CaZn_4 was also indicated by the study.

b. Calorimetry - Techniques for the combustion of aluminum in fluorine have been developed, and an initial series of combustions is in progress.

Exploratory work on the combustion of carbon in fluorine is being carried out. The work is directed toward finding a satisfactory ignition technique.

3. General Chemistry and Chemical Engineering

a. Fluid-Bed Calcination Studies in Small-Diameter Columns - Current studies in the continuously operated small-diameter ($2\frac{1}{4}$ -in.) fluid-bed calciner are concerned with the effect of deeper bed operation (15-in. depth as compared to 10-in.) on changes in particle size distribution. Runs of 5.5 and 6.0 hrs were conducted at a temperature of 445°C and an air-to-liquid volume ratio of 810, using aluminum oxide as the bed material and 1 M aluminum nitrate as feed at a rate of 18.4 ml/min. Preliminary results indicate the trends noted in the shallower bed work (see Progress Report, April 1962, ANL-6565, p. 47); namely, particle growth and increased fines formation appear to have been halted with the use of the deeper beds.

IV. PLUTONIUM RECYCLE

A study of the plutonium buildup in fast reactor blankets as a function of blanket burnup for several different reactor types has been completed using results of CYCLE calculations. The study is being oriented, where possible, to the use of a fast and a thermal reactor in which the two reactors are coupled through fuel exchange (e.g., the fuel bred in the fast reactor will be used without reprocessing as fuel for the thermal reactor). Four reactor types were studied: a 1000-liter metal core; a 1750-liter oxide core; an 875-liter carbide core; and the EBR-II. The burnup for a specified plutonium buildup is sensitive to the neutron spectrum which can be characterized by the ratios $\sigma_f^{28}/\sigma_c^{28}$ and $\sigma_c^{28}/\sigma_f^{49}$. The inner blanket of the EBR-II has a $\sigma_f^{28}/\sigma_c^{28}$ ratio equal to 0.4 while the oxide, carbide, and metal systems exhibit values between 0.09 and 0.13 for this ratio in the innermost blanket zone. This ratio decreases to 0.02 to 0.06 in the outer blanket zones of these three reactors. The $\sigma_c^{28}/\sigma_f^{49}$ ratio has a much smaller variation with blanket position and reactor type. In Table VII are given typical values of burnup (U^{238} , U^{235} , and Pu^{239}) in depleted uranium blankets (0.2% U^{235}) for specified Pu^{239} buildups and various ratios of $\sigma_f^{28}/\sigma_c^{28}$.

Table VII. Blanket Burnup as a Function of Pu^{239} Buildup

% Pu^{239} Buildup	% Blanket Burnup* for Various Values of $\sigma_f^{28}/\sigma_c^{28}$		
	0.02-0.06	0.09-0.13	0.4
1	0.08	0.16	0.4
2	0.28	0.5	0.7
3	0.56	0.86	1.2
4	0.84	1.2	1.6

*Expressed as a percentage of the original uranium atoms.

Further analysis of the data indicates that there should be only a small difference in blanket burnup (<0.1%) for any specific final enrichment ($Pu^{239} + U^{235}$) when natural uranium is used in the blanket instead of depleted uranium. The natural uranium blanket will have the slightly lower burnup for a given final enrichment.

The Pu^{240} fraction in the blanket plutonium seems to be approximately independent of the reactor type and blanket zone, and depends only upon the final plutonium enrichment. Over a range of Pu^{239} buildup from 0.5% to 4% the Pu^{240} content can be expressed by

$$\frac{N^{40}}{N^{40} + N^{49}} = \left(\frac{N^{49}}{N^{28}} - 0.004 \right)$$

where the symbols have their usual meanings.

V. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Facility Design

a. Reactor Vessel - A new beam tube layout has been developed which appears to have several advantages over the layout presented in the original concept illustrated in ANL-6451.* The advantages include: (1) a near symmetrical layout; (2) optimum nozzle spacing on the reactor vessel; and (3) similar nozzles for the tangential beam tubes at the midplane of the core. This new layout will facilitate design of the reactor vessel and will result in a substantial savings because of nozzle design.

Work on the definitive design of the reactor vessel is in progress. Certain departures from the original vessel concept have been made. A hemispherical top head will be used instead of the original flat head, in order to alleviate gasket rubbing problems. However, a welded flat head will be used at the bottom of the vessel instead of a hemispherical bolted head, to provide sufficient room for control rod drive penetrations.

b. Control Rod Drive Mechanisms - Drawings are being prepared to show the proposed control rod drive assembly and its penetration through the vessel head. The proposed drive mechanism is a combination air cylinder-motor driven device. The control blade and extension rod are connected to the piston of a double-acting air cylinder which provides the upward lifting force for the control blade and the downward force necessary for fast shutdown. A motorized lead screw assembly is attached to the head of the cylinder and is oriented parallel to the piston rod. A bracket which travels linearly as the lead screw rotates is coupled to the piston rod.

In lifting the control blade, the rod end of the cylinder is vented to the atmosphere and the piston end is pressurized to provide an upward force greater than the combined load of control blade weight and reactor pressure. When the motor is energized, the gear coupled lead screw drives the coupling bracket upward. The pressurized piston follows the moving bracket and raises the blade. The force is transmitted by contact pressure; there is no direct coupling between the lead screw drive and piston rod. Similarly, downward motion of the bracket forces the piston and blade downward.

*D. H. Lennox, et al., "Status Report on the Argonne Advanced Research Reactor," ANL-6451 (November, 1961).

For rapid insertion, an accumulator and solenoid-operated four-way valve are arranged to supply high pressure air to the cylinder to drive the rods down, with contact between the lead screw drive and piston rod being broken.

Indication of blade position is achieved through a synchro-transmitter geared directly to the lead screw. Limit switches restricting blade movement are actuated directly from the lead screw.

This type of drive unit was chosen because of the limited space available. The air cylinder compensates for control blade loading and alleviates the need for a bulky drive motor. The units as described can be mounted on the floor of the subreactor room, allowing access to all components.

c. Primary Heat Exchanger Materials - A decision has been made to use Inconel as the tubing material for all of the primary heat exchangers because of the chloride stress corrosion problem associated with the use of treated cooling tower makeup water taken from the Chicago Sanitary and Ship Canal. Cooling tower water is to be circulated directly through the shell side of the primary heat exchangers to remove the heat generated in the reactor.

Current analyses of treated canal water indicate a chloride content of 47 ppm. Based upon the required makeup rate of 800 gpm and a blowdown rate of 5%, the equilibrium chloride content of the cooling tower water would be about 1000 ppm, a condition highly conducive to causing stress corrosion cracking in austenitic stainless steel heat exchanger tubing. The situation would be aggravated by the aeration action of the cooling towers, which tends to saturate the water with oxygen and increase still further the probability of stress corrosion cracking.

The possibility of employing complete demineralization of the 800-gpm cooling tower makeup water to limit chloride content was investigated and found to be both costly and uncertain in effectiveness. Such a treatment system could not be guaranteed to remove all chlorides, and even a small amount entering through the makeup would result in intolerable equilibrium concentrations. Moreover, demineralization would have no effect on oxygen content.

The alternate solution adopted is to use Inconel tubing in heat exchangers instead of austenitic stainless steel. Inconel is considered to be completely resistant to stress corrosion cracking under AARR conditions, so that a high degree of system integrity is obtained despite the unfavorable water conditions.

d. Shielding Evaluation - An investigation has been carried out to assess the effectiveness of shielding of the primary coolant system in the event of a complete core meltdown after operation at 150 Mw. From data obtained from the fuel meltdown incident in the Westinghouse Test Reactor, it was calculated that such a meltdown would result in the release of 1.61% of the fission product inventory to the reactor coolant. Instantaneous dispersion in 10,000 gal of reactor cooling and no decay was assumed. The equilibrium fission product spectrum of Roos* was used and shows that the maximum dose rate in any area accessible to personnel during reactor operation was 5.9 r/hr penetrating the canal floor above the primary coolant cleanup system cubicle, at a point 6 ft above the 24-in. pipe returning coolant to the reactor vessel. The floor here consists of 2 ft of ordinary and 1 ft of magnetite concrete.

The dose rate above the canal floor originating from the primary cleanup resin column under meltdown conditions was also calculated. For this case it was assumed that all released fission products were adsorbed in the top 12 in. of the 28-in. diameter resin bed after 2 hr of accelerated cleanup operation. The dose rate penetrating the canal floor to a point 18 ft above the top of the column was found to be 2.5 r/hr. Here the fission product spectrum of Perkins and King** was used.

2. Research and Development

a. Core Physics Calculations - The beneficial effects of the hard neutron energy spectrum expected in the stainless steel core were further investigated to determine the recovery time after a power setback. The effective Xe^{135} cross section varies between 0.16×10^5 and 0.57×10^5 b. Therefore, with respect to xenon kinetics this reactor behaves in the same way as a thermal reactor with a flux of 10^{13} to 4×10^{13} . This follows from the fact that the xenon absorption area is a function of the microscopic cross section and the flux. Hence, the maximum δk needed to recover from xenon is 3.75%. If only 2% δk is available, typical setback times are of the order of $5\frac{1}{2}$ hr at the beginning of life and $1\frac{1}{2}$ hr at the end of life. Samarium poisoning is negligible under these conditions, since the maximum δk resulting from samarium after shutdown is less than the reactivity gained from xenon decay.

*Matt Roos, "Sources of Gamma Radiation in a Reactor Core, Part B. Reactor Technology," J. Nucl. Energy, Vol. 1, p. 99 (1959).

**J. F. Perkins and R. W. King, "Energy Release from the Decay of Fission Products," Nuclear Sci. and Eng. 4, pp. 726-46 (June, 1958).

b. Core Heat Transfer and Fluid Flow - A critical survey was initiated to investigate the heat transfer and fluid flow problems associated with the reactor core and shielding.

A hydraulic loop capable of operating in the range of flow and temperature conditions expected in AARR is near completion. The test loop is designed for investigation of flow characteristics of single sub-assemblies. One such stainless steel subassembly has been fabricated. Plans have been made for visual observation and for motion pictures to be taken of the subassembly during the test runs. In this way collapse of the plates can be correlated with flow velocities. According to calculations, collapse is not expected until velocities of 65 to 75 ft/sec are attained; these velocities are much greater than those contemplated in AARR.

B. Subcritical Reactor as a Substitute for Isotope Heat Sources

There is a requirement for SNAP-type power sources for sizes which lie between the small units powered by isotope decay heat and the large units powered by reactors. This region of interest is between 50 to 500 watts of electrical energy. A subcritical multiplying medium surrounding a strong neutron source has received preliminary study and shows promise for this application. The neutron source could be 100 mg of Cf^{252} providing approximately 3×10^{11} neutrons/sec by spontaneous emission.

The systems listed in Table VIII were considered as possible candidates for the multiplying assembly.

Table VIII. Minimum Weight Critical Systems of Various Compositions

Materials and Neutron Spectra	Total Weight (Fissionable Material + Moderator) (kg)
Pu^{239} (fast)	16
U^{235} (fast)	48
U^{235} , Li^7H Moderator (thermal) (Li^7 isotope)	15.9
U^{235} , Beryllium Moderator (thermal)	228

From the data it is seen that a Li^7H -moderated assembly with a multiplication constant (M) of 500 ($k_{\text{eff}} = 0.998$) would produce 3750 watts of thermal energy and with a thermal conversion efficiency of 5% would yield 187 watts of electrical energy. The study of the subcritical system will be continued.

C. Direct Conversion Studies

1. Thermionic Energy Conversion Diode Using a Film Boiling Liquid Metal

The conventional plasma thermocouple or thermionic energy conversion diode has a solid metal electron collector. Cesium vapor at a pressure of a few millibars or less is widely used to provide ions for electron space charge neutralization. Usually conditions are such that a monolayer of cesium coats the collector so that electrically the collector appears to be cesium. This suggests the use of a diode with a liquid metal collector material such as cesium, rubidium, or potassium and their alloys. Recent experiments at ANL have shown that a diode can be produced consisting of a hot emitter and a liquid metal collector separated by the vapor film created in film boiling. In such a diode, the metal vapor pressure can be brought in the range of a few millibars or less by, for example, controlling the pressure of an inert gas blanket which covers the film boiling liquid. If conditions in the film are appropriate for the production of plasma, a thermionic energy conversion plasma diode results.

An experimental diode was designed to operate continuously. A cross section is shown in Figure 4. It is heated by an electron gun which occupies the center section. The filament and focusing cone can be seen just above a tantalum cylinder which acts as electron emitter. The cylinder (1.45 cm diameter) is chamfered at 45 degrees and has a polished bottom (1.05 cm diameter).

Figure 4. Diode with Liquid Metal Electron Collector

The electron gun assembly is isolated by an enclosure made of tantalum and stainless steel. Cooling lines entering and leaving from the upper left carry silicone oil to a single turn cooling jacket in the stainless steel portion of the gun assembly. The tantalum portion into which the emitter is welded is made of 0.254-mm sheet. The tantalum rim is sandwiched between stainless steel and fusion welded to form a vacuum-tight closure. The liquid metal electron collector is contained in the conical copper cup located below the tantalum emitter. The silicone oil cooling jacket is shown below and the line through which liquid metal is admitted extends to the left. Metal vapor evolved from the liquid rises through the annular space outside of the electron gun assembly and condenses on copper fins attached to the outside wall. The top of the chamber which contains the liquid metal and the top of the gun assembly are cooled by water coils. They are electrically insulated from each other by a thin teflon gasket between the flanges which carry the rubber "O" ring seal. Both the gun and the liquid metal chamber are continuously pumped by a liquid nitrogen cold trap and mechanical forepump. The liquid metal chamber can be isolated and filled with a low pressure argon gas blanket. The pressure is measured by a Dubrovin gauge (range 0-26 millibars). The gas enters through an opening not shown just above the condenser fins. The experiment can be observed and pyrometer and liquid level readings made through a sapphire window located in a sight tube seen on the left.

Each experiment was done with a constant potassium inventory contained in the pool, on the fins, in the form of vapor traveling upward and droplets draining downward.

When the emitter had reached the proper temperature, gas was introduced until the pressure far exceeded the vapor pressure which potassium would have at the temperature of the copper cup. Under these conditions, the potassium in the cup was subcooled, that is, below the boiling point. Subcooling of the liquid tends to condense the vapor in the film boiling sheath causing transition boiling to occur with the accompanying intermittent electrical contact. This behavior could be observed externally by noting the shape of the current voltage characteristic of the diode which was continuously displayed on an oscilloscope. This characteristic was produced by driving the diode voltage both positive and negative with an external 60-cycle voltage source. With this oscilloscope trace as a guide gas was slowly pumped out of the diode, thus raising the boiling point, until intermittent shorting was observed to cease and give way to the normal characteristic of an energy conversion diode. This operating point measured the maximum subcooling at which film boiling could take place.

Figure 5 shows the temperature of the potassium collector (T_c), plotted against the saturation temperature of potassium (T_s); that is, the boiling point of potassium corresponding to the gas pressure.

Figure 5. Collector Temperature (T_C) vs Potassium Saturation Temperature (T_S) for Maximum Subcooling.

Data are given for two electron emitter temperatures, 2233°K and 2115°K. The diagonal line is the trace of $T_C = T_S$. The data points would have fallen very near this line if a saturated liquid had been required for film boiling to exist. Thus, the deviation of data points from the line is a measure of the subcooling that can be tolerated and still have film boiling under the conditions of temperature measurement and operation that have been described. In this case, the maximum submersion depth is 1.6 mm at $T_C = 609^\circ\text{K}$. Thus, the temperature limits are defined within which the diode could be operated with a constant potassium inventory.

It can be seen from the two sets of data points presented here that less subcooling can be tolerated for a lower emitter temperature. For the emitter at 2115°K, the maximum submersion depth is 1.8 mm at 609°K. A region of thermal instability, indicated by the dashed box, was encountered in the middle part of the curve for the emitter temperature equal to 2115°K. The temperature of the collector tended to oscillate upward or downward through the unstable region, preferring operation at either the upper or the lower end of the curve. The onset of the instability was accompanied by ripples in the vicinity of the emitter about 0.3 mm high and observation of the oscilloscope revealed that the degree of contact between the liquid and the emitter during the transition boiling was much greater than had been observed in the more stable region.

When the inventory of potassium had been reduced so that the maximum depth of submersion decreased to 0.5 mm at 606°K, greater subcooling could be tolerated as is shown in Figure 6 for emitter temperature 2150°K. Here a subcooling of up to 30°K is indicated. When the inventory of potassium was still further reduced so that the submersion was almost

zero, subcooling of the order of 100°K could be tolerated, producing a sheath of vapor which was estimated to be about 10^{-2} mm. Here the maximum subcooling was not as well defined, because apparently the separation was maintained more by gravity and less by film boiling.

Figure 6. Collector Temperature (T_C) vs Potassium Saturation Temperature (T_S) for Maximum Subcooling with Reduced Submersion.

During the run with the emitter at 2233°K , current voltage characteristics were recorded using an X-Y plotter at three different collector temperatures. These are shown in Figure 7 for collector temperatures 580, 599, 609°K . The potassium pressures of 0.59, 1.49, and 2.01 millibar are also indicated. In another run, when the collector temperature was held constant at 605°K and the emitter temperature was varied, the set of current voltage curves shown in Figure 8 was obtained. The vapor pressure which accompanied maximum subcooling is given. At these potassium pressures, current voltage characteristics of the observed shape are to be expected unless the electrode spacing is extremely small. The separation distance was not measured, but was estimated visually when the submersion was small to be of the order of 0.1 millimeter. From the shapes of the curves, this distance appears to be many mean free paths. However, at the lower pressures and intermediate emitter temperatures, the sharp knee of the ideal curve begins to appear.

In summary, the maximum subcooling which can be tolerated in the film boiling liquid metal diode operated in the manner described depends strongly on vapor pressure, strongly on depth of submersion and weakly on emitter temperature. However, the film becomes more unstable at lower emitter temperatures. The current voltage shapes show that vapor film thickness was great enough for transport losses to be apparent under all of the conditions investigated thus far with potassium.

Figure 7. Volt Ampere Characteristics for Various Collector Temperatures and Constant Emitter Temperature with Maximum Subcooling.

Figure 8. Volt Ampere Characteristics for Various Emitter Temperatures and Constant Collector Temperature with Maximum Subcooling.

VI. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal Oxidation and Ignition Studies

Isothermal oxidation experiments have been performed in the high-temperature "heat sink" reaction cell on low-concentration aluminum alloys of uranium. The "heat sink" apparatus prevents excessive self-heating of the alloy during the oxidation reaction by pressing the specimen cubes of alloy between large pieces of metal. Isothermal oxidation studies have shown that there is a strong dependence of oxidation rate at 500°C on the aluminum content. Table IX indicates the nature of this dependence:

Table IX. Dependence of Oxidation Rate of Uranium-Aluminum Alloys on Aluminum Content

Aluminum Content of Alloy (ppm)	Oxidation Rate at 500°C ($\mu\text{g O}_2/(\text{sq cm})(\text{min})$)
15 (ANL base metal)	915
34 (BMI base metal)	1510
55	2120
75	3600
1130 (1 a/o)	19800

Previously reported results (ANL-6145, page 140) showed that there was a sharp decrease in the ignition temperature of uranium upon the addition of elements such as aluminum. For 8.5-mm uranium cubes, the ignition temperature dropped from 570° to 420°C with the addition of 70 and 75 ppm aluminum, respectively. Similar results were obtained for silicon. These increased reaction rates and the decreased ignition temperatures of low-concentration aluminum alloys of uranium illustrate the importance of avoiding even traces of aluminum in uranium metal.

Studies of the ignition of zirconium are being made to determine the important factors and conditions contributing to the hazardous ignition behavior of this metal. The dependence of ignition temperatures of spherical zirconium metal powder on sample weight is being determined with helium-shielded experiments. Ignition temperatures of -225 + 400 mesh powder range from 365°C for 0.6 gram to 630°C for 0.01 gram in a $\frac{1}{4}$ -inch diameter copper crucible.

2. Metal-Water Reactions Studies

Studies of the reaction of solid uranium with steam to produce uranium dioxide and hydrogen are being made by the volumetric method. Results now indicate that the reaction is parabolic over the range

500° to 1200°C. A tentative value of 15.4 kcal/mole was obtained for the activation energy of the reaction. The reaction is a two-stage linear process at 400°C. It is now apparent that the reactions of uranium with steam and with oxygen both undergo a marked change from a linear or accelerating rate to a slower protective reaction between 400° and 500°C.

Four metal-water experiments have been conducted in TREAT. In three of these experiments, 89 w/o zirconium, 11 w/o uranium (93% enriched) alloy plate was melted in water by reactor bursts of 251, 372, and 452 megawatt seconds total energy. These plates reacted with the water to the extent of 4, 7.7, and 8.5 percent, respectively. In the fourth experiment a cylindrical pin of aluminum-10% U_3O_8 (93% enriched) was subjected to a 940 megawatt-second transient in water. A gas leak made the measurement of the extent of reaction unreliable. However, it is estimated from the hydrogen-helium ratio found in the autoclave following the run that there was greater than 8 percent reaction.

B. Fast Reactor Safety Studies

1. Experimental Meltdown Program

In-pile experiments are being performed in the TREAT reactor to study the important mechanisms producing failure in fast reactor fuel elements and subsequent meltdown product motion.

a. Clustered EBR-II Elements in the Absence of Sodium - The first four capsules from meltdown Series XXX (see Progress Report, April, 1962, ANL-6565) were received and examinations were begun during the report period. Capsules 5 and 6 were received but have not yet been opened. Each capsule contained seven elements, arranged on an EBR-II fuel pitch inside a close fitting stainless steel hexagonal tube similar to, but smaller in diameter, than the can enclosing an EBR-II subassembly. All samples included tantalum neutron absorbers to produce an axial power shape typical of a fast reactor core. Post-mortem results (see Figure 9) that have been obtained may be summarized as follows:

Capsule 1 - Maximum recorded sample temperature was 1040°C. The hexagonal can was intact. Small, nonviolent sample failures were found in the central fuel region.

Capsule 2 - Maximum recorded temperature was approximately 1400°C. The hexagonal can was not penetrated by fuel alloy. Most of the uranium fuel was found concentrated in a segment cast into a hexagonal shape inside the bottom 7 cm of the hexagonal tubing. However, no alloying between this cast slug and the surrounding can was found.

Capsule 3 - Maximum recorded temperature was 1035°C. The central element was surrounded with six stainless steel dummy elements. Hexagonal can was not penetrated. The central element had suffered violent failure in the central fuel region. Cladding of the dummy elements was extensively penetrated.

Capsule 4 - Maximum recorded temperature was approximately 1100°C. Results were similar to those of Capsule 3.

Figure 9. Post-mortem photograph taken of the opened hexagonal cans from Capsules 1-4 or Series XXX. Each can was slit longitudinally, and the two pieces set side by side for the photograph. The bottom end during the TREAT test is at the right for each sample. Note the extensive damage to the fuel elements evidenced for Capsule 2. Wires projecting from the left end of the cans are chromel-alumel thermocouple leads.

b. Meltdown Experiments on Pre-irradiated EBR-II Elements - Preparations are underway for the first meltdown series on behavior of pre-irradiated EBR-II elements under transient nuclear heating in TREAT (Series XXXI). Three specimens, irradiated in MTR to an estimated burnup of approximately 1 at-% will be run in dry opaque capsules modified for remote assembly and handling. The final modifications have been completed on the handling coffin for these experiments. The lifting sling and the bar assembly for raising the coffin have been assembled, tested, and approved. Relative burnup along the sample axis has been measured by gamma ray scanning, and sodium bond tests of the irradiated elements are in progress using the cyclograph eddy current testing unit adapted for remote operation

in a Building 301 cave. Furnace annealing of the sample elements to normalize the metallurgical condition of the fuel is partially completed.

2. Equipment Development

a. Small (Package) Sodium Loop - The design has been completed on the handling cask for the package sodium loop designed to fit into the 10-cm square by 244-cm rectangular space of a TREAT fuel element. The cask design has been approved by the Bureau of Explosives. Drawings have been submitted to the Shops for construction.

One instrumentation and control cabinet for the loop has been assembled but has not yet been checked for operation. A duplicate cabinet, for use for out-of-pile testing and operation, is under construction.

The modified fixture for the molybdenic oxide-uranium "thermite" heat source tests has been installed in the prototype test loop. Actual testing using this chemical heat source is awaiting the installation of modified pressure transducer housings.

b. Large Sodium Loop - A large sodium loop, designed to be a semi-permanent installation in TREAT is being constructed for exposure of clusters of sample elements in flowing sodium. Shipment of this equipment to Idaho has commenced. The shipment includes hardware for the stairwell in the reactor basement, equipment room crane, piping, and valves.

The loop heater specifications are now being prepared.

c. Sodium Vapor Pressure Measurement - Severe embrittlement of the tantalum leads occurred during the initial operation of the vapor pressure furnace. Necessary modifications were made to the furnace including the substitution of carbon for the tantalum. Furnace testing has been resumed and several vapor pressure determinations were made to check the apparatus. Data were taken over a range of conditions up to 10.6 atm in pressure and 1215°C in temperature.

d. Instrumentation for Measuring Transient Pressures in High Temperature Sodium - Since the maximum temperature ratings of the currently available miniature fast-response pressure current transducers are limited to the 340° to 370°C temperature range, it is necessary to protect them from the higher ambient temperatures existing in the proposed tests. Protection is accomplished by inserting a special pressure tap between the transducers and the sodium loop which provides temperature isolation without introducing undesirable pressure response characteristics. Transducer response to sinusoidal sodium pressure has been calculated using a simplified model which allows for the equivalent vibrating mass of transducer diaphragm plus fluid in the connecting tube between the loop

and transducer, but estimates must be made for the viscous damping effect of the fluid in the connecting tube. These calculations were made for a range of straight tubes in diameters from 0.159 cm to 1.23 cm and lengths from 5.08 cm to 50.8 cm, terminating in a CEC 4-316A high temperature transducer. In the most unfavorable case (0.159 cm diameter, 50.8 cm length) it was calculated that the response should be flat within 5% up to a frequency of about 50 cycles/sec. Because the model does not include provisions for detailed study of end effects of the tubes, bends in the tubes, etc., experimental testing of response characteristics for specific transducer attachments is required.

A mockup of the small sodium loop test section has been built to study transducer mounting techniques and response to temperatures up to 540°C. The original attachment assembly designed for the loop was found to be unacceptable because of (a) the thermal stresses induced in the zero shift adjustment of the transducers; and (b) diaphragm damage encountered when the sodium in contact with the diaphragm froze or melted.

A prototype gas free-piston engine for producing pressure pulses approximating a sine wave was built and tested. Frequencies up to about 75 cycles/sec were obtained before testing was terminated due to excessive cylinder wear and fatigue cracking of tubing connections to the cylinder. Fabrication has begun on a new engine having a hardened stainless steel cylinder and more rugged tubing connections. The testing program will consist of using this new engine with an air-water system at room temperature to test response of the attachment assemblies developed with the test section. In the case of the reference attachment design, which requires NaK in the standoff tube, the frequency response tests will be performed with water in place of NaK.

3. Theoretical Studies

a. Calculation of Gas Equations of State - An LGP-30 program has been written and checked by calculating gas densities as a function of pressure and temperature for gases satisfying the Beattie-Bridgeman equation of state:

$$P = \alpha \rho + \beta \rho^2 + \gamma \rho^3 + \delta \rho^4 \quad ,$$

where P is pressure, ρ is density; and the $\alpha, \beta, \gamma, \delta$ are the Beattie-Bridgeman coefficients, which are a function of temperature only. Actual densities are calculated by an iterative procedure using the formula:

$$\rho_{n+1} = 1/\alpha(P - \beta \rho_n^2 - \gamma \rho_n^3 - \delta \rho_n^4) \quad ,$$

where

$$\rho_1 = P/\alpha = P/RT \text{ (perfect gas law)} \quad ,$$

and n is the number of iterations.

Through the use of this technique it is possible to calculate densities under conditions where the perfect gas law would be less accurate. A test problem was run in which hydrogen densities were calculated for temperatures of 100°K and above. At 2440°K, one iteration was found sufficient to calculate density for one atmosphere, using a convergence criterion that the change in calculated density between successive iterations be less than or equal to 0.01%.

b. Heat Transfer Calculations of TREAT Meltdown Experiments on EBR-II Elements in Flowing Sodium - Calculations have been performed using the RE-147 "CYCLOPS" heat transfer code for studying the temperature distributions within EBR-II samples undergoing transient power excursions in TREAT. The geometry and heat transfer coefficients used as input for the code problems were those that were discussed in previous Progress Reports (ANL-6473 and ANL-6485). The output of these problems has been analyzed to determine the effects of sample enrichment, sodium coolant flow velocity, and initial excess reactivity (or time duration) of the transient power excursion.

The coolant velocities investigated ranged from 182 cm/sec (6 ft/sec) to 752 cm/sec (25 ft/sec) and values of initial excess reactivity of the power excursions ranged from 1.2% to 2.4%. All the transients were temperature terminated and axial flux shaping was not specified.

The temperature that is considered to give the most information about the initiation of failure of the sample is the temperature at the interface of the fuel and the cladding. For this reason relationships have been determined between this temperature and enrichment, coolant velocity, and excess reactivity.

In analyzing these relationships it was noted that there exists a threshold of coolant velocity above which violent failure will not occur. The behavior of this velocity threshold is such that it decreases with increasing initial excess reactivity and with increasing enrichment. For an enrichment of nearly 3% and an excess reactivity of nearly 1.5% this threshold is 450 cm/sec (15 ft/sec). This result may be compared with the experiments on uncooled samples, for which it was found that a temperature terminated excursion initiated with 1.5% k_{ex} and a 3% enriched EBR-II specimen would generate about three times the energy necessary for violent failure.

The temperature distributions determined to date can be considered applicable in time until either the sample fails or the sodium coolant begins to vaporize. When vaporization begins the vapor that is formed restricts the flow of the coolant.

Work is continuing to study the effects of enrichment, liquid coolant velocity, and initial excess reactivity on the time at which vaporization begins, and the effects of vaporization on the temperature distributions within the sample.

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