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REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

April 1961

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

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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:

October 1960	ANL-6253
November 1960	ANL-6269
December 1960	ANL-6295
January 1961	ANL-6307
February 1961	ANL-6328
March 1961	ANL-6343

	<u>Page</u>
D. Heat Engineering	57
1. Axisymmetric Free-Convection Heat Transfer Study	57
2. Hydrodynamic Instability	58
3. Measurement of Power-to-Void Function	58
4. Void and Velocity Distribution in Two-Phase Flow	58
5. Packed Bed Reactor Studies	59
6. Boiling Liquid Metal Experiments	59
7. Physical Behavior of Reactors and Reactor Systems	59
8. Dynamic Correlation Measurements of Heat Transfer and Fluid Flow Characteristics	60
E. Separations Processes	60
1. Fluidization and Fluoride Volatility Processes	60
2. General Chemistry and Chemical Engineering	62
3. Chemical-Metallurgical Process Studies	63
F. Advanced Reactor Concepts	64
1. Compact High Power Density Reactors	64
2. Fast Reactor Test Facility (FARET)	65
3. Direct Conversion Survey	66
VI. Publications	68

I. WATER COOLED REACTORS (040101)

A. EBWR

1. 100-Mw Modifications - Reboilers

a. Repairs - The repairs on No. 1 reboiler were completed early in the month, and the unit was ready for final inspection on April 11, 1961. A Laboratory representative witnessed the helium leak test of the tube welds during the final inspection. The repairs appeared to be successful up to that time although the integrity of the tubes was questionable.

Preliminary helium leak tests during the final testing period revealed numerous leaks. Attempts to locate the sources of leakage by probing with the mass spectrometer were unsuccessful due to the excessive leakage. A soap bubble test performed on the face of the tube sheet revealed that 50 to 100 tubes in the top half of the bundle were leaking. Only ten possible leaks were found in the bottom half of the bundle, which was consistent with the experience of the failures in No. 2 reboiler. The vendor agreed that the No. 1 reboiler was beyond further repair.

The tubing recently removed from No. 2 reboiler was inspected and found to be embrittled, as evidenced by jagged ends of the tubing removed from the tube sheet and cracking of the tubing upon flattening.

The vendor has agreed that both reboilers will require new tube bundles and tube sheets, and has initiated work on both units. The new tubing will be welded stainless steel rather than the seamless tubing used in the original units.

b. Metallographic Examinations

Unused Tubing - Tubing samples taken from a U-tube rejected for wrinkling in the 180-degree U-bend (approximately that within the central two-thirds portion) resisted boiling 65% HNO₃. Material from the ends to this portion of the tube are not available for tests; the location of the material used for the exploratory investigation was not determinable. The samples might have been from tube ends cut from the long tubes to form the shorter inner U hairpin bends of the bundle.

Reboiler No. 1 Tubing - Longitudinal and transverse specimens were prepared from four parts of a nonleaking tube. The samples were subjected to a two-hour exposure to 20% HNO₃ at 180°F. The locations of the samples from the accessible outer tube of the tube bundle were:

- (a) Upper leg of U-tube - end rolled into tube sheet.
- (b) Upper leg of U-tube - 3 ft from (a).
- (c) Bottom leg of U-tube - end rolled into tube sheet.
- (d) Bottom leg of U-tube - 3 ft from (c).

The photomicrographs revealed:

1) Flow lines which had been observed in virgin metal tubing were absent. This was not unexpected since it is known that the reboiler had been stress relieved at least once at 900°F for 17.5 hr.

2) A significant difference in the structure between the two ends of the tube was observed. In tubing from the top leg of the U-tube (both the rolled end and the section three feet away), grain boundaries were strongly outlined by continuous carbide networks. The sections from the bottom leg of the U-tube were completely austenitic and free from carbides. The precipitation of carbides (as chromium carbide) lowers resistance to corrosion, thus the results of the Huey tests reported in the March, 1961, Progress Report (ANL-6343) are explained. Tubing from the bottom part of the U-tube was less resistant to a 240-hr exposure to boiling 65% HNO₃ than virgin metal tubing; the metal weight loss was five times greater (2.67% vs 0.5%, respectively). The tube end which had been cold rolled prior to and succeeding the welding of the tube to the tube sheet was preferentially roughened and darker colored.

3) In addition to the presence of carbide networks, photomicrographs of the upper leg of the U-tube showed both a scattered and a band concentration of a needle-like precipitate. From its appearance and its black etching characteristic, the phase appeared to be martensite. Repolishing, re-etching, and examinations at higher magnifications (to 1600X) confirmed the presence of this phase.

4) Material in the upper leg of the U-tube showed evidence of intergranular attack. Cracks extended from the surface inward a distance of two grains (approximately 0.001 in.).

Reboiler No. 2 Tubing - Tube samples approximately 12 in. long from the end adjacent to the tube sheet were received. The most unusual characteristic of these tube sections is the total absence of the metallic "ring," i.e., the damping factor is very high.

2. 17-4 PH Investigation

A rack, pinion, and seal shaft, heat treated to condition H-1100, were tested and the wear after 6000 cycles found to be negligible.

Material for four new racks, pinions, and seal shafts has been received and solution heat treated. Four racks will be machined (two with teeth on both sides and two with teeth on one side) and heat treated according to new specifications to develop a heat treatment that will minimize the amount of straightening necessary after final heat treatment.

3. EBWR Core I Fuel Burnup

Burnup calculations have been started on Core I fuel to bring knowledge of the core inventory up to date as accurately as possible. For these calculations the reactor is divided into zones, three axial and three radial, each differing in void content, element type, and starting isotopic distribution.

Axial power distribution is obtained by an R-Z PDQ calculation. For each of the three axial regions, an X-Y TURBO code calculation is made, the axial leakage being included. The first run of the TURBO code indicated several items of input data which were in question. These have been corrected and a second run is being made.

4. Reactor Decontamination Studies

Although it has always been possible that a fuel-element failure might occur in EBWR, the chance of failure is increased with continued use of the original Core I elements. Decontamination studies are therefore being pursued with two main objectives in mind: (1) to determine quantities of radioactive material which might deposit in the steam systems of boiling water reactors as a result of fuel ruptures; and (2) to determine what methods of decontamination can be used to remove these deposited activities.

The first study is being made with a Type 304 stainless steel loop that simulates the action of a boiling water reactor. Mixed fission products are introduced into the loop to simulate a fuel rupture. Quantities and types of fission products deposited on internally mounted metal sample strips are determined, principally with the aid of a 256-channel gamma-scintillation spectrometer. In a parallel effort, studies are being made in the laboratory to find a suitable means of removing the deposited activities.

Two series of runs were made with the stainless steel Type 304 pilot plant loop. In the first series of three runs, stainless steel Type 304 sample strips were used; SAE Type 1018 mild steel strips were used in the second series of two runs. Marked variations with time were observed in the fission product content of liquid and vapor samples. For example, the content of cesium-137 in loop liquid varied by a factor of three, whereas zirconium-niobium-95 content decreased in some cases by a factor of 100.

Marked differences were also observed in the deposition characteristics of several isotopes, as a function of elevation, on both stainless steel and mild steel sample strips. For example, in a single run, the percentage of the total activity deposition attributable to cesium-137 varied from 10 percent to 50 percent at different elevations. In the same run the increase of cerium-137 deposition occurred at the expense of zirconium-niobium-95, which decreased in the elevation interval from about 80 to 30 percent.

Current laboratory experiments are being conducted on oxalic acid and citric acid solutions containing hydrogen peroxide. A gross decontamination factor of 30 was obtained on stainless steel Type 304 surfaces when a solution of 0.16 M sodium oxalate and 1 M hydrogen peroxide was used (pH adjusted to 4 with oxalic acid). The effects of several parameters, such as solution composition, pH, temperature, and time, on decontamination were studied with a solution of three molar hydrogen peroxide adjusted to a pH of 3 and containing 0.5 to 0.05 molar potassium oxalate. The decontamination of ruthenium, the only identifiable residue, was improved by decreasing the oxalate concentration.

The corrosiveness of hydrogen peroxide in both citrate and oxalic acid solutions was measured on two low-alloy steels. The addition of hydrogen peroxide was found to result in a marked decrease in the corrosion, but decreasing the pH increased the attack. The stability of oxalic acid-hydrogen peroxide solutions was found to be poor (visibly decomposed) at unadjusted pH's. Less than 10 percent change in concentration was observed in 24 hours with the pH adjusted upward to four. Peroxide concentrations remained nearly constant when a pH of four was maintained by continuous addition of oxalic acid.

5. Reactor Operation

The EBWR control rod drive components were removed from the reactor and examined (reported in March Progress Report, ANL-6343). Extremely fine surface markings were found on the 17-4 PH stainless steel racks, but it could not be determined whether they were machine finishing marks or microscopic cracks. After determining that no serious hazard could occur, even if the racks failed during reactor operation, the racks were reinstalled.

EBWR was started up on April 17, 1961. Cold criticals were run on April 17, the plant was brought to startup temperature during the night, hot criticals run on April 18, and then the plant was put on nuclear heat. The instrumentation required only minor adjustment and is working satisfactorily. A gain of a factor of 3 in chamber flux as a function of

power level has been observed. This is probably due to peaking of the flux at the spikes, and the reduction of reflector thickness between the outer fuel elements and the vessel brought about by increasing the core diameter from the nominal 4 ft to 5 ft.

Stepwise power increases were made until 20 Mwt was reached. The reactor was shut down on Friday, April 21, 1961 and cooled over the weekend to permit minor modifications and repairs. The reactor was placed in operation again the following week.

6. Core II

Specifications for fabrication of Core II (fixed rods) fuel elements have been reviewed and bid requests will be sent out in May. A preliminary design for a removable rod fuel element has been completed. A sample fuel element will be fabricated for evaluation purposes.

The possibility of designing fuel elements containing ThO_2 in depleted UO_2 for the EBR-II blanket, which after irradiation in EBR-II can be placed in EBWR without reprocessing, is being studied. From a cursory review this appears to be feasible; however, several significant problem areas have been uncovered.

B. BORAX V

1. Construction

Checkout of the electrical and process instrumentation and control system was completed. Painting of the building, pipe and equipment, fitting of shielding slabs, and a general cleanup have been finished. The reactor building and plant have been accepted by ANL (with a few minor exceptions) from the construction contractor, Arrington Construction Company.

2. Procurement, Fabrication and Installation

The supplier has not yet fabricated any satisfactory evaluation superheater fuel plates. Plans have been established for rapid inspection and testing of these plates when they are delivered.

The Laboratory is fabricating the reactor internal structures. The hold-down boxes, chimneys and shield plugs for the control rod drive nozzles have been completed and shipped. The boiling core structure and the central superheat core structure will be completed in May. The peripheral superheat core structure, the boiling fuel assembly boxes, the void and boron rods, the dummy fuel assemblies and the control rods will be finished in June. The fuel-handling periscope has been received in Idaho. All nine control rod drives are on hand and special installation tools have been fabricated.

Fabrication of the water chemistry sampling panel is complete. Fabrication of 3 boiling-fuel-rod storage racks, the reactor pit bridge, reactor-building-pits guard rails, and lifting gear for shielding slabs has been finished. A prototype of a modified boiling-fuel-rod manipulator has been modified and tested satisfactorily.

An order for the reactor vessel extension spool has been placed with the Graver Tank Company.

Partial shipment of superheater fuel plate thermocouples has been received from Aero-Research Corp. The order for the high-temperature boiling-fuel-rod thermocouples has been placed with the Continental Sensing Company.

The tanks for the reactor water demineralizer and makeup water polishing demineralizers have been received.

Calibration of temperature-measuring, indicating, and recording instruments has started.

3. Design

Detailed design of the instrumented boiling fuel assembly and pressurized terminal boxes for instrument fuel assembly leads continued. Detailed design has been completed on the steam dryer for the central superheater core, three core structure storage stands, the makeup water polishing-demineralizer system, and the fuel-storage-pit demineralizer system.

An orifice plate for the top of the control rod channels in the core structure shroud has been designed to control the flow of water around the control rods. Besides preventing the bypass of an excessive amount of cooling water around the boiling fuel elements under forced-circulation conditions, the orifice will insure that the pressure drop across the control rods is not great enough under any conditions to lift the rods out of the core.

The material from which the dowels, nuts, and studs on the core structure struts and legs is fabricated has been changed from 17-4 PH stainless steel to Type 304. The remaining 17-4 PH components (Belle-ville springs, parts of the hold-down latch, reactor vessel dowels, and control rod extension shafts) have been heat-treated at 1150°F to a hardness of Rockwell C-33.

Work is continuing on the physics data and curves for the Operating Manual. These include control rod reactivity worth, steam void reactivity worth, temperature reactivity effect, xenon reactivity effect, boric acid

reactivity effect, reactivity versus period, decay heat, samarium reactivity effect, reactivity worth of superheater flooding versus temperature, and burnup reactivity effects.

A series of RE-147 problems has been submitted to investigate the validity of the assumption (made in the analysis of the maximum accident) that all heat generated in a boiling zone during an excursion remains in the oxide fuel.

4. Development and Testing

a. Control Rods - Static corrosion tests at 600 psig saturated conditions of Boral samples clad with X8001 aluminum, simulating the former reference control rod material, were run with the following results:

(a) two unclad samples of Boral failed catastrophically during a one-week test; (b) two clad and vented samples grew in thickness by 0.025 in. after one week and by 0.033 in. after two weeks; (c) one clad and unvented sample grew 0.34 in. in thickness in two weeks, while another sample showed no growth. Because of these results, the cladding material for the Boral has been changed to $\frac{1}{16}$ -in. thick seal-welded stainless steel Type 304. An expansion space is provided at the top of each blade for evolved helium gas. Calculations of the effect of the steel on reactivity worths have been started.

b. Boiling Fuel Elements - Ultrasonic examination has been continued on fuel rods for the boiling core of Borax V. A total of 388 elements were inspected for wall thickness and integrity by resonance and pulse-echo techniques, respectively. No longitudinal defects were detected and thickness measurements varied only a few ten-thousandths of an inch around the 15-mil specified thickness.

Vibration tests in the air-water loop on a sample boiling fuel rod using a mockup fuel assembly, both with and without a center grid, continued. A horizontal jet of water impinging on the rod from the hole in the side of the fuel assembly used to measure the vibrations was found to be a contributing factor in the large amplitudes measured previously. The test equipment has been modified to reduce this error, and a new air-injection system is being fabricated in order to obtain better simulation of the actual boiling conditions within a fuel assembly.

c. Superheat Fuel Elements - The reference fuel for the superheat fuel in BORAX V consists of flat plates containing stainless steel-uranium oxide dispersion-type cores clad with stainless steel. The plates are nominally 25.250 in. x 3.665 in. x 0.030 in., and are designed with four different contents of UO_2 , depending on plate location in the reactor core. A total of 840 plates containing highly enriched UO_2 are required. In addition, 208 similar plates containing depleted UO_2 have been ordered for evaluation and practice assemblies.

The results of the evaluation of the first 24 plates showed conclusively that none of the plates examined were acceptable (see Progress Report, March, 1961, ANL-6343). The supplier has now begun fabrication of a second set of 24 evaluation plates.

The assembly of fuel plates into finished fuel elements will be done by the Laboratory. Fixtures for production assembly and vacuum furnace brazing of the fuel elements are currently being fabricated from stainless steel and Hastelloy C.

Sections from vacuum furnace brazed assemblies of stainless steel plates are now undergoing tests in superheated steam. The brazing alloy and carrier cement used in these assemblies is Coast Metals 60 and polystyrene-benzene, respectively. After three days of exposure at 540°C and 600 psi, no change was found upon visual inspection. Testing of these samples is being continued.

d. Thermocouples - Evaluation and corrosion testing continued on the gold-nickel alloy, "NIORO," proposed for brazing thermocouple sheaths in the pressurized terminal boxes.

Tests have been started to determine if any metal-atmosphere or ceramic-metal reaction occurred on some tantalum-sheathed, BeO and MgO-insulated thermocouple samples during calibration up to 4200°F.

e. Control Rod Drive - The one 17-4 PH stainless steel extension rod that has been reheat-treated, etc. (see March Progress Report) has completed approximately 1000 full-travel cycles and visually shows little or no wear or adverse effects.

Nine of the ten drive mechanisms have been shipped to Idaho for assembly with the reactor. This will expedite the installation of piping and electrical wiring required before operation. The drives will probably not be operated except for precritical checkout until all extension shafts have been replaced. The tenth mechanism is being used for extension rod testing.

f. Physics Analysis for Superheater Critical Experiment - The physics analysis of possible hazards associated with critical experiments on BORAX V superheater elements in the ZPR-VII facility has been completed. Some interesting results are:

- (1) Twelve flooded superheater elements in the cluster are nearly critical.

- (2) The heat capacity of the superheater elements is sufficient to prevent fuel plate melting even if three out of twelve fuel boxes flood instantaneously.
- (3) The chief shutdown mechanism available is the Doppler coefficient in the region surrounding the superheater elements.

5. Operations

A training program for BORAX V operators and Operations Supervisors has started. Classes, taught by the BORAX V staff, are held three mornings per week, with an hour lecture followed by on-the-job instruction and application of lecture material. Preliminary copies of the Operating Manual are being used as a text book for this course.

II. SODIUM-COOLED REACTORS (040103)

A. General Research and Development

1. ZPR-III

a. Physics Analysis - Theoretical studies have been directed at understanding the large discrepancies between experimental and theoretical critical masses for the systems containing large amounts of aluminum and stainless steel. For example, use of the 16-group cross-section set of Yiftah et al.* (using the nonconservative ν^{25} values) results in the calculated critical masses being about 19 to 22% too small for the aluminum assembly No. 23, the oxide assemblies Nos. 29 and 30, and the aluminum-stainless steel assembly No. 31. The stainless steel assembly No. 32 is calculated about 28% too small in critical mass.

By detailed consideration of the effects of the scattering resonances of aluminum and stainless steel in the energy range between about 10 kev and 1 Mev, the above discrepancies are reduced to about 11% to 14%, and 19%, respectively, in these assemblies. Because of the high-density reflectors, the effect of the re-evaluated transport cross sections upon the critical mass is largely compensated by a corresponding increase in reflector saving, so that the increase in calculated critical mass is largely due to re-evaluation of the elastic removal cross sections.

These scattering resonance considerations then place the assemblies Nos. 23, 29, 30, and 31 within the same range of calculated critical mass deviation from experiment as observed in many previous assemblies. The calculation of the stainless steel assembly No. 32, however, would have to undergo a further decrease in reactivity of about 2% k. Considering the possible error in the capture cross sections of the stainless steel components, it appears reasonable to increase the group capture cross sections of stainless steel. The calculated critical masses of the ZPR-III assemblies would then be overly reactive by less than 2 or 3% k, which may be reduced to less than 1 or 2% k by ad hoc use of the less reactive "conservative ν^{25} values" referred to by Yiftah et al.

Assembly 33 was a modification of Assembly 32, which contained 9.3% U^{235} , 63.5% steel, and 17.8% sodium (0.86 g/cc). Multigroup calculations indicated that Assembly 33 would be bigger by 5%, indicating that the sodium cross-section values are overreactive, probably in σ_{tr} . In the experimental measurements very few changes were observed as compared to Assembly 32.

The fission ratios are given in Table I for both assemblies.

*S. Yiftah et al., Fast Reactor Cross Sections, Pergamon Press (1960).

Table I. Fission Ratios for Assemblies 32 and 33

	<u>Assembly 32</u>	<u>Assembly 33</u>
σ_f^3/σ_f^5	1.51	1.51
σ_f^4/σ_f^5	0.367	0.370
σ_f^6/σ_f^5	0.117	0.118
σ_f^8/σ_f^5	0.045	0.047
σ_f^9/σ_f^5	1.20	1.21
σ_f^{40}/σ_f^5 (Counter No. 12)	0.382	0.400
Rossi- α	$5.37 \times 10^4 \text{ sec}^{-1}$	$5.46 \times 10^4 \text{ sec}^{-1}$

In a similar manner, the reactivity measurements indicate little change in the spectrum, with most measurements showing no significant change when expressed in terms of reactivity cross sections. The reactivity results for both assemblies have been given previously in ANL-6328 and ANL-6343.

Measurements on both Assemblies 32 and 33 of the variation of the reactivity values of sodium with both density and position were made. The results are given in Table II. The error in the measurements is at best $\pm 5\%$.

Table II. Reactivity Values of Sodium

<u>Radial Position</u>	<u>Mass of Sodium</u>		
(inches from center)	<u>87.6g</u>	<u>131.4g</u>	<u>175.2g</u>
<u>Assembly 32</u>			
3.078	6.5 lh	8.0 lh	11.5 lh
6.531	8.0	11.7	13.5
7.850	6.0	9.0	11.0
<u>Assembly 33</u>			
2.180	8.1	-	15.5
6.531	8.8	-	18.7
10.885	6.4	-	12.8

b. Experimental - Work on Assembly 34 started April 7, 1961. This is a large, dilute core intended to simulate a large power reactor using uranium carbide as a fuel.

This assembly contains one column of enriched uranium, two columns of graphite, two columns of depleted uranium, three columns of stainless steel, and eight columns of reduced-density aluminum per drawer. In terms of volume percent, the composition is as follows:

Material	Vol. %
U ²³⁵	4.67
U ²³⁸	10.35
C (=1.43)	10.64
Al	25.46
Stainless Steel	24.64

The ratio of carbon atoms to uranium atoms is 1.06, and the total uranium in the core is about 3 percent enriched. The critical mass of this core is 503.01 kg of U²³⁵, and the critical volume is 574.5 liters. The approximately 25 volume percent aluminum is intended to simulate approximately 50 volume percent sodium.

Central fission ratios have been measured and are given in Table III. Measurements of central reactivity coefficients are under way.

Table III. Central Fission Ratios in Assembly 34

$\sigma_{238}/\sigma_{235}$	0.03391
$\sigma_{234}/\sigma_{235}$	0.2467
$\sigma_{233}/\sigma_{235}$	1.4544
$\sigma_{236}/\sigma_{235}$	0.08035
$\sigma_{239}/\sigma_{235}$	1.0668
$\sigma_{240}/\sigma_{235}$	0.2709

Considerable mechanical trouble has been experienced with ZPR-III. The structure which supports the movable carriage drive nut broke. This allowed the drive screw to bind and made the separation of the halves difficult. The structure was repaired by welding without dismounting the movable carriage.

To reduce the weight of this loading, the outer two rows of radial blanket material were removed leaving a minimum of eight inches of blanket rather than the minimum of twelve inches initially loaded. Removal of this blanket material caused the loss of 38.4 inhours of reactivity from the assembly. Maximum weight, before unloading the blanket material, was 24.1 tons per half.

Following four days of successful operation with the repair weld, the structure again failed. Four days of operation were lost while the weld was strengthened by extending the bead further around the plate which holds the carriage drive nut. The reactor resumed operation on the last working day of the month.

2. ZPR-VI and ZPR-IX

a. Building - Construction of Building 315 is estimated to be 98.8% complete. Door bearings of the No. 4 and No. 5 cells have been replaced with new bearings of higher capacity. The manufacturer of the air-lock doors has experienced trouble with effective seals. A new design using an inflatable seal is expected to solve the problem.

On April 17, 1961, ANL took over occupancy of most of the building. Some electrical work and adjustment of the ventilating systems remain to be completed.

b. Building Tests - The program for this building includes pressure testing of the cells for structural behavior and leak rates. Because of the leaky doors, gaskets, and other defects which are still being corrected, the pressurization and measurements of leak rate and deformation have been delayed. Everything has been made ready to proceed with the testing as soon as the contractor turns the cells over to ANL.

An investigation has been made of the zero drift characteristic of the particular strain gage technique to be used in monitoring the strain imposed upon the cell walls during pressurization. The magnitude of all of the probable errors in the electrical system which could be measured was evaluated. The instrumentation and calibration is now considered completed and the method of analyzing the test has been worked out in detail.

A preliminary investigation was made of the relationships between surface temperature changes and deflections of the inner surfaces of the faces of the cells. Temperature and deflection readings were taken hourly for several days, and then during shorter periods of favorable weather conditions. The temperature difference changes between inside and outside average surface temperatures covered a range of about 40°F. This range is considered sufficient to establish the correction factors and the preliminary investigation has terminated. The instrumentation will be left in place for use during the pressurizing and strain gage testing and the leak rate test proper in case such corroborative data should be required or appear desirable. Calibration and check-out tests were conducted for the dry-bulb-wet-bulb apparatus (forced air psychrometer), and the dewcel probe and instrument to be used for the leak rate test proper.

c. Procurement - About 200-300 stainless steel matrix tubes for the ZPR-VI assembly are being received weekly. As of now, 1000 tubes have been received and accepted. Provisions are being made for the bundling and machining of clusters of these tubes for use in the ZPR-VI system. A batch of 50 aluminum tubes for the ZPR-IX matrix has recently been received from the vendor. All except two failed to meet specifications.

The source drive mounting brackets, the fuel drawer transport carriages, and several thousand of the stainless steel mockup plates for the cores to be studied in ZPR-VI, have been received. A delay on the delivery of the bed and table assemblies was requested so that the two cells could be prepared more completely for the mounting of these assemblies. It is expected that such preparations will be finished early in May. Procurement of other components is proceeding without significant delays. Assembly drawings and bills of materials are now approximately 90% complete for the ZPR-VI system, and approximately 80% complete on ZPR-IX.

d. Control Rod Drive - Two types of control rod drives are to be used for these reactors: (a) a dual-purpose type drive which will position a fuel drawer during operation or eject it on command; and (b) a blade insertion type drive which will insert a boron-filled shim rod into the core.

Bids have been received for the fabrication of up to 20 dual purpose control rod drive units. Fabrication will begin May 1, and the first completed drive is to be delivered on or about August 1, with the remainder to follow during that month. Bids are being solicited for the fabrication of 12 to 24 blade insertion drives.

e. Experimental Program and Equipment - A study was made of the feasibility of making neutron lifetime measurements by the pile noise technique in fast criticals. It appears that a fission chamber is the most efficient detector for this purpose. Detectors employing moderation could not be used, as they would destroy the desired correlations. One type of fission chamber which is available contains 1.5 gm U^{235} . Using a criterion that the signal due to pile noise be at least equal to that from detector noise over an appreciable portion of the frequency range, this detector would only give usable chamber efficiencies in cores with less than 10 kg U^{235} content. Chambers containing 1.72 gm U^{235} are now manufactured and thus would raise this limit to 14 kg. Since the fast critical cores proposed for the ZPR-VI program would have much larger U^{235} content, it is concluded that the pile noise experiment is not feasible for those cores.

In the February, 1961, Progress Report (ANL-6328, p. 12) the achievement of 13% resolution for 400 kev neutrons with the newly-constructed Perlow spectrometer was reported. It was indicated that this was not a significant improvement over the resolution obtained with an earlier model, and disappointment was expressed that the more careful control of the assembly of this counter with regard to maintaining circular symmetry had not produced a greater improvement. The difficulty has now been traced to impurities in the gas filling used during those measurements. This has been rectified and the run repeated. The resolution resulting from these additional measurements was approximately 7% for the same filling pressure and incident neutron energy. Thus, the resolution available with the Perlow counter now compares quite favorably with that of an improved version of the methane-filled proportional counter. Preparations are now being made for further testing of the Perlow spectrometer with a fast neutron spectrum.

Two new bodies for the methane recoil-type spectrometer are under construction. They have been designed to permit spectrum measurements with better spatial definition and with less volume displacement in the assemblies in which measurements might ultimately be made. One 2 in. diameter counter of this type has been modified and the counter is ready for further testing. The design of a $\frac{7}{8}$ in. diameter counter has been completed and fabrication is underway. The assembly and testing of a high purity counter filling system is now essentially complete. It will be used in connection with the methane-filled recoil spectrometer and other systems. The sodium "getter" technique, which is being tested in connection with this gas filling system, should be suitable for purification of any likely components of gas counters to be used in the future.

Circuitry for a study of pulse shapes, including a fast, low-noise amplifier, has been constructed. Resolution of the chamber pulses will be affected if the band width is not chosen to emphasize that part of the profile subsequent to the initial avalanche from all of the electrons in the ionizing track. Some pulse-shape studies will be made to determine the character of the pulse rise for electron conversion tracks from γ -ray background.

3. JUGGERNAUT

a. Construction - The JUGGERNAUT is a water-cooled and graphite-moderated reactor of the ARGONAUT type. It is to operate at power levels up to 250 kw and provide a versatile facility for nuclear research and component development.

Work on the reactor has consisted largely of trouble shooting and making corrections.

Since construction of JUGGERNAUT is substantially completed, future work will concentrate on critical experiments, facilities for research experiments, and preparations for various experimental programs. Therefore the monthly report on JUGGERNAUT will appear henceforth in Section V, Nuclear Technology and General Support, under the subheading of Experimental Reactor Physics.

b. Critical Experiments - Fabrication of boron-containing dummy fuel plates has started. The primary purpose of these plates is to provide a means of determining accurately the delayed neutron fraction, β_{eff} , of the reactor; however, use of the boron plates during full power operation is also being considered. A study has been made which indicates that no significant hazard is associated with the use of these plates in either capacity.

c. Physics Analysis - A study of the effect of shim rod motion on the fluxes within the thermal columns indicates that the gradient of the thermal flux can be reduced to zero in the vicinity of the high flux end of

the thermal column, while reducing the magnitude of the thermal flux by only 35%. This effect may prove helpful in reducing the possibility of significant error in the fission cross section and yield experiments planned for the east thermal column.

B. EBR-I

1. Core IV: Examination of Irradiated Prototype Fuel Elements

The reference fuel material for the Mark-IV core of EBR-I is cast Pu-1.25 w/o Al alloy, NaK bonded in Zircaloy-2 tubes. In order to determine the behavior of Pu-1 w/o Al alloy (the composition originally specified for the Mark IV loading) four prototype fuel elements were fabricated and placed in EBR-I in the spring of 1960. Two of the elements contained central thermocouples.

The two unthermocoupled elements were removed from the reactor after they had achieved an estimated maximum burnup near 0.1 a/o. Both elements were irradiated with central fuel temperatures near 385°C.

The fuel rod (8.5 in. long, 0.200 in. diameter) in each element was found to be in excellent condition. A photograph of one of the fuel rods is shown in Figure 1. One rod shortened 0.001 in., and the other shortened 0.004 in. No measurable changes in diameter occurred. One rod was bowed 0.018 in. over its length. One of the fuel rods had been assembled with a wrapping of perforated niobium foil. Figure 2 shows the irradiated rod before the foil was removed.

Figure 1

Typical Pu-1 w/o Al Alloy Rod after
0.1 a/o Burnup

The results from the two prototype elements indicate that Pu-1 w/o Al alloy, and in all likelihood the Pu-1.25 w/o Al alloy, will serve satisfactorily in the EBR-I Mark-IV loading to at least a central temperature of 385°C and a burnup of 0.1 a/o.

Figure 2

Irradiated Pu-1 w/o Al Alloy Rod with Perforated Niobium Foil Wrapping

2. Fabrication of Core IV Fuel Elements

A total of 555 fuel and blanket rod assemblies are required for the EBR-I Core IV loading. This includes 420 fuel rods, 120 blanket rods, 10 fuel thermocouple rods and 5 blanket thermocouple rods. An excess over the required number is being fabricated in order to provide evaluation material and replacements for process rejects and assemblies that may not pass final nondestructive tests.

a. Casting - A total of 816 rough cut fuel slugs have been stored prior to upsetting the ribs and final machining operations. Additional material was machined for samples for physical testing. A series of 25 rough cut slugs have been upset in 0.235 in. diameter coining dies to evaluate the upsetting technique. A load of $4\frac{1}{4}$ tons (185,000 psi) was required to produce adequate ribs on the fuel slugs. The slug body diameter increased to 0.238 in. average. A slight increase in density occurred in some of the slugs which was recovered by annealing at 400°C for a period of three hours. The average slug weight was 23.34 grams and the average plutonium-239 content was 21.89 grams. Because the weight of plutonium is slightly too high, it is necessary to reduce the diameter of the fuel slugs to 0.232 in. $\pm$ 0.001 in. Tests are under way to determine the best method of reducing the casting diameter the required three to four mils.

The problem of frothing in the injection casting melts (see Progress Report, February, 1961) has been found to be related to the amount of corrosion products on the plutonium charge material. Careful removal of all loose corrosion products from the surfaces of the plutonium prior to casting has minimized the problem. Recovery of useable metal from residual heels in tantalum liners has been accomplished by inverting the tantalum liner in a yttrium oxide-coated, bottom-pour magnesia crucible in an induction-heated furnace. Upon melting the useable metal drips through the crucible pouring hole into a copper mold and the more viscous oxidized metal remains in the magnesia crucible. The tantalum liner is recovered for reuse.

Production Zircaloy fuel tubes have not been received for the jacketing of the blanket and fuel elements. However, tests have continued and techniques have been improved for assembly, welding, decontamination, NaK filling, bond testing, and leak detection of the Core IV fuel elements. A live center tail stock fixture was constructed for the welding lathe to improve the girth weld joint alignment. Tests were run on the ultrasonic decontamination tank and a good scrubbing action was produced in the cleaning zone. The pressure leak detection apparatus was completed during the month and evaluation tests are under way.

b. Blanket Uranium - Punches and dies for upsetting upper, middle and lower blanket slugs have been received and tested. Dies for the lower slugs were reworked and 20 lower blanket slugs of good quality were produced after the modification. Revision of dies for upper and middle slugs is progressing.

c. Hardware - Eddy current inspection of Zircaloy-2 jacket tubing is complete. Of 2912 feet received from the vendor, 712 feet produced an eddy current trace similar to traces from material containing fissure type tube wall defects deeper than 0.002 in. Of the remaining 2200 feet containing defects equivalent to 0.003 in. deep fissures, 381 feet were accepted for the blanket loading and 1819 feet were tested with equipment of greater sensitivity. This final nondestructive test indicated that 1100 feet of the tubing was relatively free of defects. The remaining 719 feet of rejected material contain unidentified defects which resemble large inclusions or possibly microcracks. All sound accepted tubing will be used for plutonium fuel rods.

Two finish-machined blanket rod jacket assemblies showed no visible evidence of rib wire weld failure after 60 thermal cycles. However, there was evidence of an average 0.0003 in. decrease in the 0.347 in. diameter which includes jacket tube and one rib wire. The assemblies were cycled through a minimum of 160°C by immersion of the closed end to a depth of 5 in. in 300°C oil followed by air cooling. Each complete cycle averaged 50 minutes in length.

All the required rod tips, restrainer springs and 64% of the stainless steel extension rods for EBR-I Core IV have been received.

A 4500 foot order of commercially fabricated 0.062 in. diameter Zircaloy-2 wire was received. This wire was purchased as a backup effort for Zircaloy-3 rib wire being fabricated at ANL. Destructive and nondestructive tests have shown this Zircaloy-2 wire to be of poorer quality than the material fabricated by the Laboratory which is currently being used in production of jacket assemblies.

Efforts are being made to fabricate 0.080 in. diameter Zircaloy-2 tubing for instrumented assemblies from ANL-extruded material, as opposed to reducing larger size tubing obtained from commercial vendors. It is expected that in this way all phases of the process from ingot to finish tube will be known and can be controlled. Because of the smaller size of the initial tube which can be extruded, 0.410 in. O.D. x 0.320 in. I.D., the amount of cold work necessary to produce the desired size tubing is reduced. Results obtained from tubes fabricated from the first extrusion have indicated numerous small cracks on the inside surface of the tube; however, wall thickness uniformity and concentricity were generally good. The depth and number of I.D. cracks represent a significant improvement over commercially available material or material previously fabricated by cold drawing.

C. EBR-II

1. Construction

The approximate status of the construction contracts as of April 18, 1961, was as follows:

Building	% Completion
Power Plant (Package 2)	100
Reactor Plant (Package 2)	100
Sodium Boiler Plant (Package 3)	94
Fuel Cycle Facility (Package 3)	87

a. Sodium Boiler Plant - The Progress Report for January (ANL-6307) noted the Laboratory's concern that adequate care was not being exercised by the Contractor to assure the degree of cleanliness required in the secondary sodium and steam systems. In order to substantiate this concern more thoroughly, a series of X-ray pictures were made by the Laboratory of selected portions of the secondary sodium piping of which installation was complete. Figure 3 is an X-ray photograph of a nut-bolt-washer-wire assembly found within the piping by this investigation. This assembly, commonly termed a "mouse," normally holds a circular disc of rubber, leather, asbestos, cardboard, etc., and is used during welding operations to effect a temporary internal pipe closure in order that an inert gas atmosphere can be maintained on the root pass of the weld. After completion of the weld the "mouse" is withdrawn from the pipe by use of the wire attached thereto. Figure 4 indicates typical

foreign material - probably slag from welding or cutting operations - found in a number of places. In order to estimate the density of material shown on the X-ray negatives, known penetrometers are incorporated in each X-ray negative. These are visible on the left side of Figures 3 and 4.

Figure 3

Foreign Object in Secondary Sodium Piping

As a result of this preliminary inquiry, a more extensive investigation is now in progress. This will consist of a complete X-ray examination of all secondary sodium piping of 4 in. O.D. or greater. Approximately 20% of the piping has been examined to date and the following additional foreign objects identified: (1) a second "mouse"; (2) a 6-in. section of weld rod; (3) a complete weld rod; (4) a dense object approximately 1 in. by $\frac{3}{8}$ in.; (5) a less dense object about 2 in. by $\frac{1}{4}$ in. - probably sheet metal; and (6) more slag and scale of the type shown in Figure 4. Corrective action will be taken after the results of the complete survey are available.

The contractor is required to (1) submit a mill ladle and check analysis of all piping material; (2) identify all piping by repetitive marking of ASTM designation and heat number; and (3) submit chemical analysis of field samples from each lot of piping. The first incomplete report of chemical analyses of field samples was recently submitted and contained three samples of $2\frac{1}{4}\%$ chromium-1% molybdenum piping.

Figure 4

Welding Slag and Scale in Secondary Sodium Piping

Two of the samples were of thin-walled sodium piping and one sample was of heavy-walled steam piping. All three chemical analyses identified the three samples to be SAE-4130 material rather than the $2\frac{1}{4}$ % chromium-1% molybdenum material specified. These samples were taken by the contractor from pipe now installed in the system and the certified analyses were supplied by the contractor.

The contractor has been notified that these analyses are not acceptable. The contractor has been requested to (1) submit the required number of sample analysis; (2) identify piping from which samples of this first submittal was obtained and repeat the sampling and chemical analysis procedure; and (3) take the necessary action to assure the Laboratory that all piping material meets the requirements of the specifications.

2. Installation of Equipment - Package 4

Installation work within the reactor plant falls into two general categories: (a) mechanical, comprising approximately 70% of the total; and (b) electrical, approximately 30%. For convenience in scheduling and supervising the work in the field, the mechanical work is divided into 43 major jobs, or packets, and the electrical into 5 packets. Work on 42 of the mechanical packets and all 5 of the electrical packets has been started. To date, approximately 95% of the total mechanical work and 95% of the electrical work has been completed.

It is anticipated that during the month of May the Package 4 Contractor will vacate the Reactor Plant premises to permit ANL to commence the preparatory work required for starting of the dummy loading of the reactor and the subsequent approach to dry critical. After completion of the dry critical experiments, the Contractor will return to the Reactor Plant to complete the Package 4 work within the Plant (flexible seal assemblies, heat exchanger, remainder of the secondary system sodium piping, etc.).

Several photographs pertinent to the installation work accomplished are presented in this report as described below. Considerable progress in these areas has been made since the photographs were taken.

a. Gripper and Hold-down Mechanism - This device has been described in the March Progress Report (ANL-6343). Figure 5 is an over-all view of the gripper and hold-down mechanism taken during erection. This picture shows the main support columns to the left of the gripper and hold-down, and the support platform directly above. The latter is fastened to the upper structure of the reactor vessel cover lifting mechanism which can be seen to the right of the gripper and hold-down. The plastic-wrapped units surrounded by the upper structure of the reactor vessel cover lifting mechanism are the control rod drives. Figure 6 shows the hold-down shaft being raised preparatory to installation.

Figure 5

Installation of fuel gripper and hold-down mechanism on the small rotating plug in the Reactor Building

Figure 6

Installation of fuel gripper and hold-down mechanism on the small rotating plug in the Reactor Building

b. Storage Rack Mechanism - This unit has been described in a number of previous Progress Reports (e.g., ANL-6343). Figure 7 shows the shield plug for the storage rack mechanism prior to installation. Figure 8 shows the storage rack itself setting on the primary tank bottom prior to installation of the storage rack plug. This photo was taken looking directly into the primary tank through the "T" nozzle. Figure 9 shows the storage rack as it was uncrated on the floor of the reactor building. Clearly shown is the perforated outer wall designed to permit convective cooling of both the storage rack structure and the contained reactor subassemblies.

Figure 7

Storage basket plug on the crane in the Reactor Building

Figure 8

Storage basket in the primary tank in the Reactor Building. Photo taken through the nozzle

Figure 9

Closeup view of storage basket in the Reactor Building

3. Engineering

a. Electrical Work - The electrical packets for dry critical experiments have been completed and issued. A survey has been completed on the emergency electrical loads on the 400-kw diesel generator. Based on the results it was decided to connect a surplus 200-kw diesel generator to carry the requirements of the fuel cycle facility. This extra capacity will enlarge the over-all emergency power system and permit additional loads if necessary.

Packets covering the auto-lockout and time delayed application of the emergency loads will be completed prior to wet critical tests.

Engineering for seven additional electrical packets to be completed prior to wet critical are essentially complete. It is estimated that 770 man-days electrical labor will be required to complete these packets.

b. Operating Manual - The electrical staff is devoting approximately 50% of its effort to the preparation of the operating manual. Rough drafts and drawings have been completed on several sections. The instrumentation staff has started the primary system instrumentation writeup for the Reactor Operating Manual.

c. Shield Design - Calculations were made to estimate the shielding required for the 3-in. diameter duct up through the EBR-II sodium surveillance periscope. The tube extending above the shielding on top of the reactor will require a 2-in. thickness of lead around it to reduce the gamma-ray dose at the surface to about 2 mr/hr. A plug at the end is needed to provide a surrounding shield equivalent to 5 in. of lead.

4. Procurement

The March Monthly Progress Report (ANL-6343) estimated that the sodium-to-sodium heat exchanger would be delivered in April. It is now estimated that shipment will be made in late May, 1961, which will be satisfactory.

The Fuel Handling Console was completed at the Datex Company plant and subjected to exhaustive tests by members of the Laboratory's staff prior to shipment. Both the Console and the Numerical Position Control Equipment are being installed and the equipment is expected to be ready for tests on or about May 8.

5. Component Development - Instrumentation

a. Automatic Flux Control - An automatic flux control system is being designed for EBR-II which will control reactor power to $\pm 1\%$ from

10% to 100% of full power. An on-off control system will be used in order to utilize the control motors and power contactors provided for manual operation.

A phase-space analysis has been applied to the simulated reactor control system to determine an optimum control switching boundary. The possible application of a predictive type of on-off control system was also considered. Phase portraits were taken of the system with six groups of delayed neutrons in the reactor kinetics and with simplified second order and third order approximations. The following conclusions were drawn:

(1) The system as designed meets the performance specifications with a minor amount of switching action under the input condition of small step disturbances. Changing the switching boundary will not appreciably improve the system performance.

(2) For a future full-range power demand control system, a predictive type automatic control system should prove to be very useful.

b. Control Rod Reactivity Generator - A sinusoidal reactivity generator and wave analyzer are being designed to measure the transfer of EBR-II. The reactivity generator will utilize a specially designed rotary mechanism which will fit into a control rod location. A velocity servo accurate to 1%, over a 10 to 1 speed ratio, will be used in combination with gear changes to cover the frequency range from 0.01 to 100 radians/sec. The wave analyzer used to measure amplitude and phase shift will be similar to the one used on EBWR.

The final model of the nulling circuit has been checked for spontaneous oscillations when measuring low input currents. Preliminary measurements have shown that the input amplifier will deliver 100 volts d-c at the output with an input of 4×10^{-7} ampere. It is stable for all combinations of resistors from 10^4 to 10^9 ohms and equivalent input cable capacitance from 10^{-9} to 10^{-8} farads. The frequency and phase response of the amplifier is flat up to 2 cps with any input resistance from 10^4 to 10^7 ohms. With a 10^8 ohm resistor and a gain of 40 the phase shift is 15 degrees at 2.5 cps, and 50 degrees with 10^9 ohms and a gain of 4. More accurate data will be taken with the high resistance values.

The mechanical layout of the oscillator rod, which is to be calibrated during the dry critical experiments, has been essentially completed. The rod will have a total stroke of 8 in., and is designed to be compatible with the existing reactor fuel handling equipment. Preliminary design requirements have been established for the motor and mechanism for the linear oscillator.

c. Startup Instrumentation - The startup instrumentation system was demonstrated to operate satisfactorily under laboratory conditions, and all equipment was shipped to Idaho on April 21, 1961, with the exception of the thimbles which should be completed by May 15, 1961.

d. Neutron Source - A third stainless steel clad antimony rod has been ordered, and will be irradiated to provide a low level source. The tantalum tubing required for cladding the antimony for wet operation is not expected to be delivered until late May.

e. Automatic Control System - An analysis of the two sodium loops for determination of the transport (distance-velocity) delays was completed. An analysis of the steam loop for transport delays was initiated. An analysis of the heat exchanger and superheater for the variance of the heat transfer coefficients at 10% and 100% flow was completed. A transfer function for the reactor for 50% power was calculated. The 10% and 100% power transfer functions had been previously computed. It is planned to use the 10%, 50%, and 100% power transfer functions of the reactor to establish a non-linear analog computer model. This model will be used to study the transient characteristics of the reactor for automatic control purposes.

6. Component Development - Steam Generators

Two evaporators are being modified for use as superheaters. The modification will consist of installing a central core within each tube in order to increase the steam velocity (see March Progress Report, ANL-6343). The design of core tubes and methods of fabrication and installation are being studied. The core tube will probably consist of a thin-walled, $\frac{13}{16}$ in. O.D. carbon steel tube. The tube will be concentrically located in the 1.065 in. I.D. evaporator tube by dimpling the core tube at intervals of about 24 in. along its length. The dimples will be effected by local distortion of the tube diameter such that the dimpled core tube will just have sufficient radial clearance to be inserted into the evaporator tube. Upon proper positioning of the core tube within the evaporator tube, its ends will be closed by a similar procedure thereby positioning the core tube axially and eliminating bypass steam flow through the core tube.

An attempt to vacuum melt and cast a ferrous alloy to be used as weld rod for making the tube-to-tube sheet welds on the modified evaporators is being made. To date, four castings have been made and all four had to be rejected. Two were unacceptable because of porosity, one because of low manganese analysis, and one because of high carbon analysis.

7. Component Development - Fuel Reprocessing Facilities

a. Fuel Cycle Facility Design and Testing - Much of the equipment for use in the Air and Argon Cells has been procured. Some of the equipment is still undergoing design. Invitations to bid for the shielding window shutters

have been sent out. These shutters will cover the inside of the windows when they are not in use in order to reduce exposure to gamma radiation. A degassing furnace will be used to degas process equipment items before they are put into the Argon Cell. This furnace has been received from the manufacturer and will be tested soon. The Model 8 manipulators to be used in the Air Cell have been shipped to Idaho by the manufacturer.

Eight toggle switch boxes for remote control of cranes and manipulators were built by Clement Electric Company. These were tested and returned to the manufacturer for corrections when defects were found. They have now been retested and six were found to be satisfactory. The other two require minor corrections.

b. Development of Remote Fabrication Equipment - The design of the refabrication equipment for the EBR-II Fuel Cycle Plant is estimated to be 76% complete. Designs were completed on a combination torquing and lifting tool, and on a calibrating device for the data processing and readout equipment which will simulate the signal output from the inspection machines. Checking, final design changes and detailing were completed on the fuel pin processing and inspection machines. Drawings were completed on the depalleting machine and an assembly drawing was made on the fuel pin and tube assembly machine. Both these latter devices perform functions which might be more effectively done by master slave manipulators; therefore, it was decided that procurement of these items should be held up pending reinvestigation of the availability of suitable master slave manipulators.

The plug-in pin processing equipment modules and feeder devices were mocked up and tested for manipulator disassembly and reassembly for feeder action, for sealing of pneumatic connections, and conductance of the electrical contacts. Minor changes in the design of the modules were required as a result of these tests.

When crucibles are heated in the melt refining furnace, large temperature gradients between the crucible wall and bottom may result in thermal stresses that are sufficiently severe to crack the crucible. Efforts are being made to minimize this possible cause of crucible failure. Temperature gradients in the crucible are being measured and new patterns of applying power to the furnace are being studied. When the power is applied in small steps, it is found that the temperature gradients in the crucible are substantially reduced.

Irradiation tests of materials which may be used in the Argon Cell continue. A grease compounded from Standard Oil NRRG-335 and Shell APL radiation-resistant greases was found to be satisfactory after a gamma dose of 1×10^9 rad.

Experimental confirmation of the self-heating calculations was started. The apparatus for this includes dummy fuel pin castings employing Kanthal electric resistance heaters in Vycor glass molds, and electrically heated dummy fuel elements which may be arranged in the various process configurations. The experimental apparatus may be used for bench testing or placed in prototype equipment to study self-heating effects and cooling requirements. The wiring of these dummy specimens is approximately 70% completed.

8. Process Development

a. Melt Refining Process Technology - In the melt refining process for EBR-II core fuel, off-gases will be pumped through a filter into a shielded storage tank where they will be held until conditions are suitable for the stack discharge of the gases to the atmosphere. Some observations of the behavior of off-gas activities were made in connection with melt refining experiments using highly irradiated EBR-II type fuel pins. The principal activity observed in the off-gases, other than krypton and xenon, was iodine-131.

The final experiment was completed on the evolution of radiokrypton and radioxenon during the heating of highly irradiated EBR-II fuel pins. The fuel was irradiated to 0.4 percent total atom burnup and cooled 35 days. The evolution of xenon-133 was observed as the temperature was increased in steps from 256° to 1144°C. The bulk of the xenon was released between 750° and 1070°C. The melting of the pin was complete at the latter temperature. The results obtained in this experiment agreed substantially with the results of the two experiments reported previously (see Progress Report, November, 1960, ANL-6269). The apparatus for the present experiment was modified to permit accurate measurement of the xenon-133 activity released at the lower temperatures of the experimental temperature range. The release of the noble gases at these lower temperatures is of process interest because irradiated EBR-II fuel pins may become heated to these temperatures (about 250°C) by fission product decay heat. The percentage P of total xenon-133 activity released in time t (minutes) may be represented by the empirical equation $P = a + bt$. The constants for the equation at several temperatures are as follows:

Temp (C)	a	b
256	0.2×10^{-3}	0.66×10^{-4}
379	6.1×10^{-3}	1.74×10^{-4}
619	3.7×10^{-3}	2.73×10^{-4}
749	0.2×10^{-3}	3.86×10^{-4}

The behaviors of iron, nickel, and chromium in the melt refining and skull recovery processes are of interest, since there is a possibility that small amounts of stainless steel from the pin jackets may enter the processing cycle. All three constituents were found to behave similarly to the noble metal fission products, both in the melt refining and the skull recovery processes.

b. Processing of Melt Refining Skulls - Work was continued on the development of the skull reclamation process. The behavior of iodine in the first step of this process, namely, the oxidation of the crucible skull material at 800°C to free it from the crucible, is currently under study. In a recent experiment employing a lightly irradiated fission-uranium alloy (5.3×10^{13} fissions/gram), 30 percent of the iodine remained in the skull after melt refining. About one-fifth of this 30 percent vaporized during the skull oxidation. The remaining four-fifths remained in the oxide or was present in the crucible walls. All iodine was contained in the oxidation furnace. A Fiberfrax plug over the crucible was an effective iodine trap. The distribution of iodine throughout the system, however, may vary with irradiation level.

The second step of the skull reclamation process is a noble metal extraction step in which noble metals are extracted from the fission oxide by suspending the oxide in a molten salt flux and contacting the resulting slurry with zinc. Two runs with slightly irradiated fission oxide showed rapid extraction (within one hour) of about 75 percent of the ruthenium, but little additional extraction with further contacting. This may be accounted for by the difficulty of extracting ruthenium from the coarse particles of oxide which settle to the bottom of the vessel.

In another series of experiments, it was found that the extent of removal of noble metals from fission oxide in a zinc-flux system was not only dependent on particle size but also on the temperature of the system. The degree of noble metal extraction increased as the temperature was raised from 650° to 750°C in the case of both fine (-325 mesh) and coarse (-14 +25 mesh) particles. The degree of extraction was found to increase as the particle size was reduced in runs made at 650° and 750°C.

The third step of the skull reclamation process consists of reducing the uranium oxide with a magnesium-zinc solution in the presence of a flux. As the reduction occurs, the uranium transfers to the metal phase. After the reaction, the flux and metal phases are separated. In one method of phase separation (pressure siphoning), heels of each phase are allowed to remain in the vessel. Since the reduction and noble metal extraction steps may alternate in the same vessel, there has been concern that the uranium and magnesium present in the reduction metal heel would lead to high uranium losses in the zinc used for the succeeding noble

metal extraction step. It was shown, however, that uranium is readily oxidized into the flux phase during the course of the oxidation-reduction reactions which take place during the noble metal extraction step. Uranium loss in the noble metal extraction step may, therefore, be easily avoided.

In studies of the reduction of fissium skull oxide by zinc-magnesium solutions in the presence of a salt flux, it was found that a minimum final concentration of 2.4 weight percent magnesium in the zinc phase was required for complete reduction. It was also observed that a minimum of about 50 percent of the cation concentration in the flux must be magnesium ion to produce complete reduction of fissium skull oxide. The flux components used in these studies were mainly calcium chloride, magnesium chloride, and magnesium fluoride.

The reduction step is followed by successive precipitations of a uranium-zinc intermetallic from a zinc-rich phase and uranium metal from a magnesium-rich phase with supernatant solution removals after each precipitation. Finally, retorting is carried out to vaporize residual solvent metals and recover a uranium metal product. A number of retorting runs have been made employing the above sequence of steps. In general, about 80 percent of the uranium was easily removed from a tantalum crucible in the form of agglomerates up to 1-inch in diameter; the remaining 20 percent firmly adhered to the tantalum. This adherence of uranium to tantalum has not been overcome; consequently, plans are being made to test beryllia as a retorting crucible.

c. Plutonium Recovery Processes - Further preliminary results were obtained on potential methods of separating plutonium, uranium, and fission products from the second EBR-II core fuel. The solubilities of cerium in zinc-calcium solutions containing from 23 to 100 weight percent zinc were obtained as functions of temperature to investigate the possibility of separating rare earths from plutonium and uranium by extraction with zinc-calcium alloy. Preparations are currently being made to study the distribution of cerium between plutonium and calcium.

A correlation of previous results and scouting studies indicates that selective reduction of fissium oxides by high magnesium-zinc alloys may provide a separation of cerium from uranium and plutonium. This information is encouraging inasmuch as the plutonium-rare earth separation appears to be the most difficult aspect of a second EBR-II core process.

d. Fused Salt Studies - Equipment is being constructed for phase diagram and spectrophotometric studies of fused salt systems. Experiments on the chlorination of uranium and fissium oxides to render them soluble in fused salts have been successful.

The chlorination and the subsequent reduction of the uranium tetrachloride that was formed was carried out in a sodium chloride-potassium chloride flux in the following way. U_3O_8 , suspended in the flux, was converted at 800°C to uranyl chloride by means of a chlorine-carbon monoxide mixture. Zinc was added to reduce the uranyl chloride to uranium dioxide. The flux phase was separated from the metal phase and was treated again with a chlorine-carbon monoxide mixture. The uranium dioxide was thereby converted to uranium tetrachloride. The addition of zinc and magnesium to the uranium tetrachloride-flux mixture reduced the uranium to the metal which precipitated out of the flux phase.

e. Materials and Equipment Evaluation - In 100-hour corrosion tests at 850°C , there was no detectable corrosion of tungsten or a 10 percent tungsten-tantalum alloy by a 5 percent magnesium-zinc alloy in the presence of several halide fluxes. The corrosiveness of halide fluxes to tantalum was significantly reduced by drying the fluxes. With close control of impurities, tantalum may prove to be a satisfactory construction material.

Appreciable difficulty was encountered in long-term (100-hr) runs which were made to study the stabilities of uranium-zinc-magnesium solutions in the presence of flux. The flux-metal mixtures were contained in graphite crucibles. The difficulty was caused by the considerable evaporation of zinc from the system. Nearly 250 grams of zinc vaporized from two molten metal-flux systems maintained at 850°C in essentially closed ATJ graphite crucibles. There is some indication that appreciable loss occurred by diffusion through the graphite walls.

Construction has been completed of a large-scale cadmium distillation unit which will enable engineering scale demonstrations of distillation and various liquid metal handling operations. After a final safety review of equipment and operating procedures, experimentation will be started.

9. Fuel Development and Fabrication - Core I

With the assembly of the three boron carbide safety elements, the production of EBR-II Core I has been completed.

a. Fuel Shipment - Preparations are underway to ship the first core loading to the Idaho site. Two different types of containers will be used. One type, approved by the Laboratory's Criticality Hazards Control Committee, will be used to ship all of the enriched subassemblies. A second type will be used for shipping the natural or depleted uranium subassemblies.

b. Fuel Prototype Irradiation - Erratic thermocouple behavior was observed in the irradiation rig in CP-5 and the assembly was removed from the reactor. It is suspected that the base weld on the capsule developed a leak permitting the high pressure helium (17 atm) to enter the capsule bond sodium. Upward movement of the gas through the bond would cause destruction of the element because of the high temperatures produced as a consequence.

10. Core II Fuel Development

a. Fast Reactor Fuel Jacket Development - Fuel jacket material for future fast reactors must be superior to stainless steel because of the desired higher operating temperatures. Several niobium and vanadium base alloys possess some of the required properties at temperatures in excess of 900°C. Requirements for these alloys are compatibility with both sodium coolant and fuel, good fabricability and weldability, and high strength and low creep at elevated temperatures. Vanadium base alloys have been developed by the Armour Research Foundation for fabrication development work at ANL, and niobium base alloys have been developed for ANL by Battelle. Commercial suppliers have been contacted to supply tubing, or base material which can be fabricated into tubing, of high strength alloys when available.

An attempt to draw 20 feet of 0.189 in. OD x 0.013 in. wall molybdenum tubing down to 0.176 in. OD, was not very successful. The tubing was of extremely poor quality. Cracks, holes and seams were visible to the naked eye. After sectioning to remove the defects, the tubes were stress relieved at 1000°C for one hour in vacuo. After pickling, the tubes were lubricated and hot drawn over a mandrel at 500°C. To prevent cracking and wrinkling of the tube while removing the mandrel, the tube was inserted in a 0.180 in. ID steel support tube. All finished tubing was tested with a helium mass spectrometer leak detector. A total of 2.5 feet of satisfactory tubing was accepted for jacketing irradiation test specimens.

A sample of Nb-4 w/o V alloy bar received in the as-forged condition was annealed at 1500°C for 1 hour. The hardness decreased from Rockwell-C 30-35 to Rockwell-B 90-93. Attempts are being made to anneal an additional sample of Nb-8 w/o V alloy in a similar manner. The remaining portion of the bars will then be annealed prior to boring and drawing into tubing.

The V-5 w/o Ti-20 w/o Nb alloy laminated sheet material from ARF was arc melted and cast in the shape of a square button. The button fractured during cold rolling. The material was then remelted and jacketed in stainless steel preparatory to hot rolling.

A strip of Nb-2.37 w/o Cr alloy received from BMI measuring $\frac{7}{8}$ in. x 4 in. x 0.051 in. was cold rolled 50%. The strip was then trimmed to a width of 0.803 in. and annealed in vacuo for one hour at 1225°C. The hardness decreased from 73 to 66 on the Rockwell-N scale. The strip was then sandblasted prior to forming it into a tube for welding. The strip was very brittle and fractured during the initial forming operation. A small section of unannealed strip, (cold rolled 50% to 0.025 in.), was formed into a tube without difficulty. Samples of these strips are being analyzed and examined metallographically to determine whether impurities, change in structure or a ductile-brittle transition caused the loss in ductility during annealing.

b. Irradiation Testing of Refractory Fuel Jackets - The U-20 w/o Pu-10 w/o Fs alloy fuel intended for future use in EBR-II fuel has been shown in previous studies to swell catastrophically at temperatures exceeding 400°C. However, EBR-II operating conditions require fuel operating temperatures on the order of 650°C. An additional potentially serious situation presented by the use of the plutonium alloy fuel is the fact that it has been shown to form a eutectic with stainless steel at 550°C. This temperature happens to coincide almost exactly with the expected fuel jacket temperature during normal operation of EBR-II.

It is therefore apparent that the stainless steel jackets being used for the first core of EBR-II cannot be used for the U-Pu-Fs alloy fuel. Since the refractory metals as a class have been shown to be compatible with U-Pu-Fs alloy at elevated temperatures, and since alloys can be made of these metals which are much stronger at elevated temperatures than stainless steel, it appears that the use of refractory alloy tubing for the second core of EBR-II may simultaneously provide solutions to the problems of fuel-clad compatibility and restriction of fuel swelling by jacket restraint.

An extensive irradiation program is being developed to determine by direct experiment the relative ability of various alloy jackets to restrain high temperature swelling in U-Pu-Fs alloy and to avoid fuel-clad alloy reactions. The first six specimens have been placed under irradiation in the CP-5 reactor in a controlled temperature capsule. The central fuel temperature in all specimens is 590°C, and clad temperatures are near 480°C. The cladding materials in the first capsule include niobium, niobium-0.75 w/o zirconium alloy, vanadium, Inconel X, and 304 stainless steel. Specimens with the Inconel and stainless steel claddings are wrapped with a 0.0005 in. thick vanadium foil to prevent fuel-clad alloying reactions.

11. EBR-II Physics

Some preliminary efforts have been devoted to the neutronics of a plutonium-fueled EBR-II. The work is based on the systems suggested by the Core II Development Program. A survey of existing cross-section data for the pertinent materials suggests that considerable effort must be devoted to generating multigroup constants which may be confidently used for the detailed design of such a core loading. Such work is also necessary to specify the neutronic uncertainties inherent in the core design. In reviewing the kinetic aspects it is recognized that modifications may be required on either control rod composition and/or control rod drive speed for a wholly or partially plutonium-fueled EBR-II.

The upper and lower axial blankets of the EBR-II contain about 30% depleted uranium. Less than 3% of both power and plutonium production are from these regions. There is some serious question about the advisability of reprocessing the spent axial blanket elements whose removal is dictated by fuel alloy burnup considerations. Therefore, the criticality of the EBR-II (First Loading) was compared with a similar system containing stainless steel instead of depleted uranium in the axial reflector. Few-group two-dimensional diffusion theory calculations indicate that the stainless steel axial reflector tends to increase the reactivity of the system by not more than $3.2\% \Delta k/k$. This would be reflected by a reduction in the critical mass of not more than 6.5% or 13% depending on whether core alloy enrichment or core size, respectively, is altered. The efficiency of the axial blankets might also be enhanced by the introduction of suitable moderator materials.

III. STUDIES AND EVALUATIONS (040116)

A. Fast Breeder Reactor Technology Review

This study is a review and evaluation of the status of the technology of fast breeder reactors designed for central station power applications. A review of the literature and existing research and development programs has been carried out in an effort to identify those regions of technology which need development and to determine the degree of improvement necessary to produce power at an acceptable cost.

B. Nuclear Superheat Studies

Direct and indirect cycle superheated systems were investigated using the following basic parameters:

Fuel surface temperature:	680°C (1250°F)
Steam temperature:	540°C (1050°F)
Temperature rise in heat exchanger for the direct cycle:	40°C
Heat generation per unit length of fuel:	10-100 w/cm
Fuel rod diameter:	0.65-2 cm
Power density in fuel:	1-1000 kw/liter
Heat transfer coefficient:	0.06-6 w/(cm ²)(°C)

It was found that a liquid metal-cooled reactor has a ten-fold advantage in fuel power density over a steam-cooled reactor (340 kw/liter and 33 kw/liter fuel). Applying modifying factors for the different coolants, there appears a five-fold advantage in specific power for the sodium-cooled indirect superheating system over the direct steam-cooled reactor system.

IV. REACTOR SAFETY (040117)

A. Thermal Reactor Safety Studies

1. Fuel Coolant Chemical Reactions

Knowledge of the nature and extent of chemical reactions with nuclear reactor core metals that may occur in pressurized water or steam is essential to safe operation of reactors. The principal laboratory procedure uses a condenser discharge to provide almost instantaneous heating and melting of metal wire in water or steam. The energy input to the wire indicates reaction temperature; the transient pressure measures reaction rate; light emission indicates time-temperature behavior; hydrogen generated gives extent of reaction; and particle size of the residue indicates the surface area exposed to reaction. A second method consists of heating the metal inductively and then subjecting it to a steam pulse to induce a metal-steam reaction.

The reaction of stainless steel with uranium oxides is being studied by use of differential thermal analysis.

Studies of the kinetics of metal-water reactions under reactor incident conditions are being made in the TREAT reactor.

A series of zirconium runs in room temperature water in two sizes of reaction cells and with and without added inert gas showed that the results were largely independent of these variables. It was, therefore, necessary to devise a modified theory of the mechanism by which the rate of diffusion of water vapor through the hydrogen barrier depends on water temperature. Further studies on the analog computer indicated that a very reasonable fit to the experimental results in room temperature water could be obtained using just half of the theoretical diffusion rate.

Studies of the computed results for the two-step reaction between zirconium and water revealed a very reasonable explanation for the explosive pressure rises obtained under certain conditions. Computer studies showed that a high Nusselt number was required to achieve the rapid rates. The high rates were always associated with 25 percent or more total reaction. The residue from explosive runs in room temperature water had average particle diameters less than 500μ . These facts suggested that the particles were being driven through the water at high velocity by the evolving hydrogen. Examination of high speed motion pictures indicated the presence of streaks in explosive runs. The streaks were presumably caused by rapid particle motion. Computations indicated the existence of an initial particle size of 500μ in room temperature water and about 1000μ in heated water for the rapid self-sustained reaction.

A new method of studying metal-water reaction was undertaken. It is a small-scale laboratory program in which steam is passed over levitated metal spheres and is, therefore, similar to a method demonstrated by L. Epstein and co-workers at the Vallecitos Laboratory. The program was initiated because of the failure of the condenser discharge method when applied to aluminum. A preliminary experiment has shown that a molten aluminum sphere could be suspended and heated to 1600°C by an electromagnetic field.

Studies of the aluminum-water reaction by the pressure-pulse method are continuing. Runs at 1000° and 1200°C and contact times from 0.1 to 1000 seconds have been completed. Results indicate that a cubic rate law may be applicable. A series of runs with molten uranium at 1200°C has begun. Preliminary results show that uranium is considerably more reactive than aluminum.

Size distributions of the particles produced by the in-pile meltdowns of stainless steel-urania fuel plates gave the following results by sieve-screen analysis:

Table IV. Particle Size Distribution Produced by Meltdown of SS-Urania Fuel Plates

Particle size range, mils	% of particles by weight in size range	
	495 mw-sec burst	369 mw-sec burst
0 - 1.5	39.8	6.3
1.5 - 3	12.4	2.7
3 - 6	18.9	6.3
6 - 12	17.1	7.4
12 - 28	9.3	10.8
28 - 64	2.5	42.3
64 - 132	0.0	24.2

It is evident that the more energetic reactor burst, which gave more metal-water reaction, also produced a greater percentage of fine particles.

An evaluation was made to determine the relative contribution of chemical energy release to nuclear energy release during reactor transients which result in reactions of molten uranium with water. Based on the data obtained from TREAT the following results were obtained:

Table V. Relative Contributions of Chemical and Nuclear to Total Energy Release

<u>Fission Energy calories/gram U</u>	<u>% U-H₂O reaction</u>	<u>Chemical Energy calories/gram U</u>	<u>(Chemical/nuclear) energy ratio</u>
50	0.4	2.4	0.048
100	2.4	14	0.140
150	5.0	30	0.200
200	8.9	53	0.265
300	23.8	142	0.473
450	50.0	298	0.662

The results show that over the range of fission energy inputs studied thus far, the ratio of chemical to nuclear heat is linearly dependent on the specific energy input of the reactor burst.

The study has continued on the reactions occurring between aluminum or stainless steel and UO_2 or U_3O_8 in cermet fuel pins. The method of differential thermal analysis (DTA) was used. A very mild exothermic reaction was found to occur between aluminum and UO_2 or U_3O_8 at about 900°C for 50-50 mixtures. Reaction products included UAl_2 , UAl_3 and UAl_4 which were identified by X-ray diffraction. It was concluded that the reactions were not violent or dangerous.

2. Kinetics of Oxidation and Ignition of Reactor Materials

Studies are being made of the oxidation and ignition kinetics of the metals uranium, zirconium, and plutonium in order to provide information leading to an understanding of the reactions. This knowledge should make it possible to minimize the hazards associated with handling these nuclear reactor materials. Isothermal oxidation on microscope stage, shielded ignition, burning curves, rate of propagation of burning foil, and burning temperatures are the techniques being used. In the continuing study of ignition and burning of uranium, zirconium, and plutonium, more emphasis is being placed on the burning process. Burning propagation rate studies provide a useful tool to observe the effects of many variables. The effect of the presence of halogenated hydrocarbons on the burning of uranium foil in air is being investigated.

Studies of the relationship between isothermal oxidation rates and burning curve ignition temperatures are continuing. Calculations of the specific area dependence of ignition temperatures based on oxidation only in the first stage and with transition to second stage were made. The calculated results differ from experimental results and further study is required.

A series of burning curve ignition temperature measurements in air were made with uranium foil squares of two thicknesses (one-half and five mils). Studies were made with stacks of one, two, four, and eight foil squares. The ignition temperature was 45°C lower for two five-mil foils than for one foil. Only a small additional decrease occurred with four and eight foils.

An optical pyrometer suitable for measurement of both burning propagation velocity and burning temperature has been constructed and calibrated. The instrument is currently being used to examine the effect of halogenated hydrocarbons on the burning of uranium foil in air. Combustions have been measured in which the burning temperature of five-mil uranium foil was lowered by over three hundred degrees and the burning velocity roughly halved by the addition of 20 percent CF_3Cl to air. No combustion of this uranium foil would occur in air containing 12 percent CHF_2Cl .

B. Fast Reactor Safety Studies

1. Theoretical Studies

a. Study of Ratio of Fundamental to Second Order Flux Harmonics for Oscillator Experiments - A report has been written presenting theoretical expressions for the amplitude ratio, $\underline{a}$, of second order harmonic to fundamental flux amplitudes for oscillator experiments with varying amplitudes of reactivity insertion. These equations are applicable to any reactor, provided that the linear frequency response is known up to a frequency equal to twice the maximum non-linear frequency of interest. The derivation is only valid for reactivity insertion considerably below prompt critical and is probably most useful for reactors which demonstrate a peaking in the load power transfer function. As an illustration of the method, it has been applied to a model of EBR-II and $\underline{a}$ is evaluated for zero and full power over the frequency range of 0.001 to 10 radians/sec. The model represents the effect of unrestrained thermal expansion of fuel and structure as well as coolant expansion. No bowing of fuel rods has been considered and the core is assumed to expand radially according to the expansion coefficient of steel at the coolant temperature.

b. Theoretical Studies of Destructive Nuclear Excursions - Results of a theoretical investigation of features of hypothetical nuclear bursts in fast reactors have been assembled into a report. The original Bethe-Tait method has been improved, and a number of exact calculations (using the Ax-1 IBM-704 computer program) have been made and compared with the results of the Bethe-Tait method. The basic Bethe-Tait assumptions were as follows:

1. Perturbation theory treatment is adequate for reactivity reduction.
2. Wave propagation during heat generation may be neglected.
3. Heat generation may be approximated by assuming an exponential heat generation until the reactivity is reduced to prompt critical, and neglecting the heat produced thereafter.

An improvement was obtained over the original method by discarding the last assumption, and increased yields were calculated.

Numerical calculations using Ax-1 yield even more energy release, but show considerably lower peak pressures. Investigations of the sensitivity of energy yield to parameters in the equation of state have led to scaling laws based on measuring time in units of the initial prompt period and the proportionality of excess reactivity to displacements.

c. Coupled and Fast Safety Studies (Comparison of the Safety of Coupled Fast-Thermal and All-Fast Systems) - A study has been started to investigate the safety characteristics of the coupled fast-thermal system compared to the all-fast system. The study will consider only nuclear accidents and power systems and not the special hazards associated with critical facilities. It is expected that the full spectrum of accidents to which such systems are subject will be examined. In particular, it is not intended to restrict the investigation to consideration of only the "maximum" accident. A final purpose of the investigation is that of serving as a survey of the calculational techniques available in the study of fast reactor safety.

2. Core Meltdown Studies: TREAT Program

In-pile meltdown experiments are being performed in the TREAT reactor in order to survey types of fast reactor fuel element failure and the associated movement of fuel element materials, as well as to determine the mechanisms producing such phenomena.

a. EBR-II, Mark I Dry Samples in Opaque Containers - The remaining three elements of 3% enrichment subjected to an axial "chopped cosine" power profile were examined. One sample was heated in a reactor excursion initiated with a 110 millisecond period, and having an energy release about 130% greater than that sufficient to cause extensive failure in the central part of the element. The last two samples received constant power reactor transients of about 2.5 second duration. All three samples showed results consistent with those reported in the March Progress Report (ANL-6343). Failure occurred in the central portion of the pins; fuel alloy was expelled upwards from the bottom part and downward from the top part of the elements.

b. Development of Samples with Internal Thermocouples - A series of three temperature-limited TREAT transient experiments has been performed on a natural enrichment EBR-II element into which had been cast a tantalum sheathed, tantalum-molybdenum thermocouple 0.114 cm diameter. Maximum temperatures reached in the three experiments were about 460°C, 790°C, and 1000°C respectively. Internal thermocouple readings were in reasonable agreement with those of chromel-alumel thermocouples welded to the element cladding. Two more natural enrichment elements with similar internal thermocouples have been assembled, and appear to be satisfactory. Plans are being made to use them in additional in-pile tests.

c. EBR-II, Mark I Elements in Stagnant Sodium - The four 3% enriched EBR-II samples run in stagnant sodium during the previous report period are being examined. One sample, which had received a transient calculated to reach the threshold of failure, was found to be intact but warped. The remaining three elements were found to have undergone some degree of melting. It was estimated that one should have reached the failure threshold and two should have suffered extensive failure. Their autoclaves have not yet been cut open for inspection. Study of the thermocouple recordings from the sheathed sample thermocouples used in the series indicates that the thermocouples placed nominally in the fuel-cladding bond region read temperatures more typical of the fuel than the cladding, apparently because grooves in the fuel which hold the 0.150 cm diameter sheathed thermocouples produce an extensive perturbation of the heat transfer from the 0.366 cm diameter fuel rods. The records of the thermocouples inserted in a hole in the top of the sample are consistent with temperatures to be expected from the location, but indicate that a better fuel-thermocouple thermal bond is necessary. Both sample thermocouple techniques produced temperature peaks with negligible time lags, within the accuracy of calculations of sample temperatures.

3. Component Development

a. Transparent Meltdown Facility - Their first meltdown experiments were conducted using the new transparent meltdown facility. Four 6% enriched EBR-II samples were run. All four TREAT transients were initiated with 240 millisecond periods and were clipped to yield pre-set values of reactor energy release. In addition, one preliminary transient was performed in which the reactor was scrammed early and the sample reached a temperature of about 780°C. The first two experiments were run with metal covers over the outer windows. It was found that the windows provided adequate containment; thus, the remaining two experiments were performed without cover plates. After each meltdown experiment, the fused silica window covering the inner Zircaloy capsule was found to be liberally spattered with fuel alloy. Three of the inner windows were fogged on the inside by sodium vapor.

Figure 10 is a photograph of the fuel pin obtained with a still camera after the first transient test without a metal window cover. The transparent assembly was in position in the reactor viewing slot when this picture was obtained. High speed color motion pictures were taken of the two experiments run without window covers. Failure of the two samples was clearly marked in the films by release of a cloud of sodium vapor. In agreement with previous results on similar samples run in opaque dry capsules, the release of sodium appeared to occur around the circumference of the sample, forming a cylindrical cloud rather than a "jet" or "streamer" characteristic of a spot failure. Ejection of fuel alloy was obscured by the sodium, and will be studied from still prints made from the film. The appearance and growth of the sodium vapor cloud was seen clearly before it reached the window of the inner capsule and coated it.

Figure 10

Actual Photograph of the Meltdown of an EBR-II Fuel Pin Taken
in the Transparent Meltdown Facility in the TREAT Reactor

Fission products carried by the capsule purge gas were successfully contained in liquid-nitrogen-cooled charcoal traps during and immediately after the transient tests. One day after transient test the major containment in the gas stream was Xe^{135} , which was allowed to decay in the transparent capsule assembly prior to the removal of the sample holder from the assembly. Experience with these first meltdown tests using transparent capsule assemblies indicates that one test per day is the maximum rate at which this type experiment can be run.

The large amount of stainless steel in the transparent capsule assembly caused considerable change in the reactor flux distribution, worth of control rods, and nuclear instrument calibrations. The worth of the control rods in the north half of the core was increased, while the worth of those in the south half (in which the assembly was inserted) was decreased.

b. Small Sodium Loop - Fabrication drawings of the small sodium loop are essentially completed. In addition, the shop fabrication is nearly 65% complete. Mechanical assembly of the four units has been started and the basic structures are on hand. It is planned to do the following pre-experimental tests to determine the safety characteristics of the loop. First, a series of cold shock tests will be performed wherein a predetermined number of squibs of known stoichiometric capability will be fired to produce an internal pressure similar to the most pessimistic value expected. This procedure will test the weak components of the loop. The next tests will be performed using the available thermite-U-Mo-O₂ specimens manufactured by Nuclear Metals, Inc. The units will be fired under sodium for the conditions of full flow, one-eighth flow, and stagnant. Time rate and energy release measurements will be made and measured parameters will be compared with calculated values.

c. Large TREAT Sodium Loop - Calculations were made to estimate pressure pulse and thermal stresses in the large TREAT sodium loop. The reference design has been modified for a concentric test section rather than the U-tube originally specified. Layout and flow diagram drawings are being prepared.

4. Reaction Rate of Uranium and Stainless Steel

Safety considerations for EBR-II, Core I operation require information concerning the rate of penetration of Type 304 stainless steel jacketing by molten fuel. To obtain this information a technique has been developed based on immersing 304 stainless steel capsules of desired wall thickness into the fuel. The capsule contains an insulated wire which is shorted when molten alloy breaks through the wall thus giving an indication on a high speed recorder. Penetration studies of molten uranium in the temperature range 1150 to 1350°C and molten uranium-5 w/o fission from 1100 to 1350°C have been made using 0.010 in. and 0.040 in. wall capsules.

As can be seen in Figure 11 the penetration by uranium is consistently more rapid than that of the U-5 w/o Fs. The maximum observed penetration rate for both occurs at 1150°C where it is about 15 mils/sec for uranium and 12 mils/sec for U-5 w/o Fs. In U-5 w/o Fs the rate increases from 1100 to 1150°C as would be expected. However, just above 1150°C something happens which drastically reduces the penetration rate, after which the rate increases slowly from 1187 to 1350°C. Uranium reacts similarly except that no data were taken below 1150°C because the melting point of uranium is 1132°C whereas the liquidus of U-5 w/o Fs is about 1080°C.

It was first thought that this rate inversion could be caused by the formation of an oxide film on 304 stainless steel just above 1150°C. However, Figure 11 indicates that U-5 w/o Fs penetration took place at the

same rate through the 10 mil wall as it did through the 40 mil wall. Also a plot of the data showing time to penetrate 10 and 40 mils extrapolates through zero (Figure 12) indicating that any film that may form does not significantly retard penetration.

Figure 11

Penetration Rates of Uranium and Uranium-Fissium Alloy
through Stainless Steel as a Function of Temperature

Figure 12

Penetration Time of Uranium and Uranium-Fissium Alloy
as a Function of Thickness of Stainless Steel

Another possible explanation for this slower penetration with increasing temperature was suggested by the fact that Type 304 stainless steel is austenitic at normal operating temperatures but begins to transform to high temperature ferrite at about the temperature level of interest. If ferrite could be shown to have greater resistance to penetration by molten fuel than austenite it would help to explain these anomalous results. Armco iron was selected as a material to test this hypothesis since it is known to be austenitic throughout this temperature range. Armco iron capsules were tested from 1100 to 1350°C in U-5 w/o Fs, and an inversion similar to that observed with 304 stainless steel was found. Therefore, the phase transformation hypothesis does not appear to be correct.

The formation of a high melting compound which might form a barrier is now considered to be the most likely cause for the observed slowdown in penetration rate. The high iron end of the U-Fe diagram indicates the existence of UFe_2 which melts at 1235°C and which forms a eutectic with iron melting at 1080°C. The U-Ni phase diagram has a similar compound.

V. NUCLEAR TECHNOLOGY AND GENERAL SUPPORT (040400)

A. Applied Nuclear and Reactor Physics

1. Experimental

a. $\bar{\nu}$ Measurements* - An experimental study of the dependence of the quantity $\bar{\nu}(U^{235})$ on the incident neutron energy is continuing. The actual measurements yield the ratio $\bar{\nu}(U^{235})/\bar{\nu}(Cf^{252})$. At an incident neutron energy of 1.6 Mev this ratio is now known to an accuracy of better than 1%. Small corrections due to the asymmetry of the neutron detection system must be applied to the measurements. The ratio $\bar{\nu}(U^{235})/\bar{\nu}(Cf^{252})$ also will be measured at incident neutron energies of 100, 500, and 1000 kev. The combined data will result in a much better knowledge of the energy dependence of $\bar{\nu}(U^{235})$ than now available. The incident neutron energy range is of considerable interest in fast reactor design and is a region throughout which uncertainty has existed.

Equipment suitable for similar measurements of $\bar{\nu}(E)$ for natural thorium and U^{238} has been fabricated. The actual measurements await only the availability of accelerator time.

b. Probabilities and Energetics in Fast Neutron Induced Fission - Equipment for detailed studies of the energy distribution of the fragments resulting from fission has been assembled and tested. The instrumentation utilizes solid state detectors rather than the more cumbersome fission chambers. Problems under investigation are:

- (1) The dependence of fission mass yield on incident neutron energy.
- (2) Fission cross sections of very intensely alpha active materials.
- (3) The relationship between fragment mass asymmetry and angular isotropy.

These measurements contribute to the theoretical understanding of the fission process necessary for the ultimate accurate prediction of such properties as fission cross sections.

c. Elastic and Inelastic Scattering of Fast Neutrons - Initial use was made of the millimicrosecond bunching system in experimental studies of elastic scattering of 560 kev neutrons from carbon. The high intensity of the burst makes it possible to accumulate significant data very rapidly.

* $\bar{\nu}$ is defined as the number of prompt neutrons emitted per fission.

Knowledge of the carbon cross section is not only of use directly in many applications in the reactor field but also is important as a standard against which important nuclear parameters are measured. It is for the latter reason that this problem is being carried out. Many of our previous measurements of inelastic scattering cross sections of fertile materials are relative to carbon, and data on its scattering cross section are contradictory.

d. High Conversion Critical Experiments - Supplementary information on Hi-C hazards was discussed with the Reactor Hazards Evaluation Branch representatives at the AEC headquarters. As a consequence of this meeting, the plutonium-beryllium neutron source holder is being replaced by a cadmium-covered holder made from cadmium-plated stainless steel. The new holder includes expansion space in its shank to aid in containment in the event of source rupture during a reactor accident. The cadmium cover should help shield the source from leakage neutrons from the reactor if an accident occurs while the startup source is present in the bottom reflector. The effective source strength for startup use is not greatly affected by the cadmium cover.

Study of the available information on the release of gaseous fission products from UO_2 indicated that some dispersion into the reactor cell would occur after an accident initiated by a 3% or larger positive reactivity step. Although it is believed that little or no leakage to the outside atmosphere is likely, the release of all the gaseous fission products given off by the UO_2 would produce a radioactive cloud sufficient to expose personnel at the site boundary to 0.2 to 0.3 R following a 3% k_{excess} accident.

A method for measuring the ratio of capture in U^{238} to fission in U^{235} has been investigated. The two principal problems involved in this proposed procedure are: the measurement of the weak activity of Np^{239} in a 3 wt-% enriched foil by means of the coincidence technique, and the determination of the background due to fission products in these coincidence measurements. The results of some exploratory experiments described below are encouraging.

Foils irradiated in a 1.5×10^9 thermal flux in CP-5 for 10 minutes gave a rate of 5000 coincidences in a subsequent 10 minute interval. The channels of the two single channel pulse height analyzers were adjusted to cover the entire 106 kev plus k-X-ray peak of the Np^{239} spectrum. NaI(Tl) crystals 5 inches in diameter were used as detectors. In the Hi-C experiment an exposure of $10^8 \text{ n/cm}^2/\text{sec}$ for a similar irradiation time is considered a maximum. Thus, measurements must be based on no more than a few hundred coincidences per foil irradiated. It is also probable that the Hi-C measurements will have to be made with crystals of 2 inch diameter. This will reduce the efficiency of coincidence counting by a factor of about 2.5. To get reasonable statistics, the counting time must be extended to be considerably more than 10 minutes. However, during long counting

times the equipment cannot be considered stable. Because the final results depend strongly on the stability of the counting equipment, it is important to provide for additional stabilization of the spectrometers.

In order to keep the background due to fission products low, it is advantageous to use the 5 in. crystals since they give a pulse-height spectrum in which the Compton component due to γ -rays from fission products is smaller than it is in spectra obtained with smaller crystals. However, if the smaller crystals have to be used, it is believed that the background can be determined with sufficient accuracy. A new type of source holder which will reduce the number of false coincidences due to scattering of radiation from one crystal to the other is being constructed.

e. Proposed Determination of β_{eff} - Consideration has been given to the possibility of obtaining a value of the effective delayed neutron fraction from a pile noise lifetime measurement. This would provide an alternate to the fluctuation integration method, but it would not avoid the need for an independent measurement of fission rate.

The determination would be performed as follows. The spectral density of the current fluctuations from the chamber may be written as

$$A + \frac{B}{1 + \omega^2/\alpha^2}$$

Here the first term is the chamber noise component and the second is the pile noise component. α is the Rossi alpha and ω is the frequency in radians per second. The parameters A, B, and α are obtained from a least squares fit to the observed spectrum. According to a previously developed theory [C. E. Cohn, Nuc. Sci. Eng. 7, 472 (1960)]

$$B = \frac{2Q^2 \epsilon^2 n \beta^2}{\ell^*} \frac{(\overline{\nu^2} - \overline{\nu})}{\overline{\nu}}$$

The average DC chamber current I is given by

$$I = \frac{\epsilon Q n}{\ell^*}$$

Here Q is the charge transferred in the chamber per neutron absorbed, ϵ is the fraction of neutrons disappearing by absorption or leakage which are detected in the chamber, n is the total number of neutrons in the reactor, and ℓ^* is the prompt neutron lifetime. Calculating the quantity B/I^2 from the latter two equations gives

$$\frac{B}{I^2} = \frac{2 \beta^2}{(n/\ell^*)} \frac{\overline{\nu^2} - \overline{\nu}}{\overline{\nu}}$$

Now (n/ℓ^*) is equal to $\bar{\nu}$ times the number of fissions per second. Thus, β may be obtained absolutely.

Compared to the fluctuation integration method, this scheme has the advantage that the data are obtained much faster and are less affected by extraneous reactor drifts. Accordingly, the pile noise equipment is being modified to permit measurement of the DC chamber current during a run. It will also be necessary to have an absolute calibration of the analyzer, so that the spectral density of the chamber current fluctuations can be obtained absolutely. This could be done using a random noise generator associated with the PACE analog computer.

f. Measurement of $\bar{\nu}$ for Thermal Fission of U^{235} - Since the February Progress Report (ANL-6328, pp. 57-58), the additional equipment required for the main experiments at the CP-5 research reactor has been constructed. The beam collimator, slowing-down sphere, and associated plumbing have been installed at the thermal column. The monitor fission chambers have been inserted on the face of the reactor along the beam hole and the performance tests of these and their associated circuitry have been satisfactory. Observation of the counting rate as a function of thermal column shutter position has been made, indicating that for the shutter opening likely to be used a slope of about 3% per inch of radial displacement exists for each monitor. Thus, a repositioning error of $\frac{1}{8}$ in. would be an acceptable maximum.

The coincidence counter for the manganese solution has been developed, assembled, and tested. The required solution can be poured into the annulus through tubing which penetrates its shield. Accurate volumetric determination and easy drainage and flushing have been achieved in the design. Difficulties with intermittent oscillation of the β -channel amplifier has resulted in its replacement by a different model, and preamplifiers will be added to the Cerenkov counters to insure that operation with the new amplifier is conducted on the flat portion of its gain-bandwidth curve. A measurement of background in the 4 in. by 5 in. sodium iodide crystal, which is the γ -sensitive device in the β - γ coincidence detector system, has indicated a level of about 0.7 counts per second for a wide window at the 0.85 Mev peak. This is a factor of 5 or so improvement over that existing at the previous location of the counter, and furthermore is not subject to variation due to reactor operation in the vicinity.

g. ATSR Auto-Rod Performance - The ATSR is equipped with an automatic reactivity compensation system which balances the reactivity effect due to an inserted sample by displacement of a very weak control rod. In preparation for resonance integral determinations with the ATSR, a study of the auto-rod control system has been started. The first steps have been to obtain performance data for the individual components. The performance tests indicated that the system is unconditionally stable, but that a substantial increase in gain may be necessary to obtain satisfactory transient response.

Where Milne-Thompson required data at intervals of 0.001 to obtain an error of 2 in the eighth significant digit, (using ordinary rational interpolation) for $\tan 1.5685$, the osculatory algorithm gave a maximum error of 3 in the eighth significant figure over the entire range $1.27 \leq x \leq 1.57$, with data for the three points 1.37, 1.47, and 1.57. The algorithm may also be used as an extremely rapidly convergent method of solving algebraic and transcendental equations. The algorithm was described in a paper presented at the meeting of the Society for Industrial and Applied Mathematics at Monterey, California, on 20 April, 1961.

On invitation from the program committee, a paper, "Problem Oriented Languages without a Compiler" was presented to the annual meeting of POOL, the LGP-30 Users Organization. In this paper, the value of problem-oriented languages such as Algol for communication between analysts, programmers, and scientists, was discussed. The experience at Argonne, where an untrained undergraduate summer student was able to produce acceptable programs written in Algol after approximately ten hours of instruction, was described. The extra flexibility obtainable by hand coding, where any desired arithmetic system can be used, was also stressed.

B. Reactor Fuels Development

All work on ceramic fuels and on properties of metals and alloys was suspended during April because of the move into the Fuels Technology Center, Building 212.

1. Corrosion of Sintered Aluminum Powder Product Tubing

Short lengths of impact extruded tubing made from "modified" A288 powder have been supplied by ALCOA for corrosion testing. They report yield strengths of about 8,800 psi at 315°C, intermediate between their former "as-atomized" and "milled" tubing values. These tubes have been corrosion tested at 350°C and 260°C for approximately 6 weeks. Their general appearance and weight changes are normal for resistant alloys, but microblisters were formed, particularly at 260°C.

The tubing produced by Armour Research Foundation (bridge type die) showed evidence of increased attack (after 35 days at 290°C and 41 days at 360°C) at the junction where the metal streams joined.

Closures of the powder product tubing made by Atomics International using a silver eutectic bonding process have not been adequately corrosion resistant. The eutectic bond followed by "motor arc" welding of the end has produced a strong, corrosion resistant closure. Corrosion testing of these tubes is continuing.

2. Irradiation Studies

a. Examination of Irradiated PuC and UC-20 w/o PuC - The irradiation behavior of PuC is of interest because of the potential use of this fuel in small, high temperature fast reactors. Since UC has been shown to possess excellent high temperature irradiation properties, there is some basis for the hope that PuC will also have high temperature irradiation characteristics that are an improvement over the poor performance of most plutonium alloys. UC-PuC mixtures are of interest because they combine the good qualities of carbide fuels with the capability of internal breeding in a fast plutonium-fueled reactor.

In order to explore the irradiation behavior of plutonium carbide fuels, preliminary irradiations were made in EBR-I on the nominal compositions PuC and UC-20 w/o PuC. The irradiations were made in a fast reactor because of the "blackness" of PuC in a thermal spectrum. The PuC specimens were arc-cast. The UC-PuC specimens were made both by arc melting and by pressing and sintering. The specimens were 0.300 in. in diameter and 1.00 in. long.

The first group of arc-cast samples, consisting of four PuC and four UC-20 w/o PuC specimens, have been examined after an estimated maximum metal atom burnup of 0.1 a/o at maximum central temperatures near 430°C.

All except one of the PuC specimens fractured during irradiation. The amount of cracking may be related to the carbon content. Two castings which had a carbon content near 4.9 w/o were fractured somewhat more extensively than two specimens with carbon contents near 5.3 w/o.

Dimensional changes were negligible. Three of the four specimens decreased in density, with a maximum decrease of 1.44%. The remaining specimen showed an apparent increase in density of 0.44%. A typical specimen is shown after irradiation in Figure 13.

Figure 13

Typical Cast PuC Specimen after 0.1% Metal Atom Burnup at a Central Temperature of 430°C.

Two of the four UC-20 w/o PuC specimens fractured during irradiation. However, the cracking which did occur was less severe than for the PuC specimens. In general, the UC-PuC specimens with lower

carbon content (~4.8 w/o) were less crack-sensitive than the specimens with higher carbon content (~5.6 w/o). Dimensional changes were negligible. All specimens decreased in density, with the maximum change being -1.57%. A typical irradiated specimen is shown in Figure 14.

Figure 14

Typical Cast UC-20 w/o PuC Specimen
after 0.1% Metal Atom Burnup at a Central
Temperature of 430°C.

b. Examination of Irradiated Metal-Fibered Thoria-Urania Pellets -

The poor thermal conductivity of oxide fuel has limited more than any other single factor the heat rating which may safely be tolerated in oxide fuel elements. In an attempt to improve the thermal conductivity of ThO_2 - UO_2 fuels, ThO_2 pellets containing 10, 30, and 50 w/o UO_2 and 10 w/o of either molybdenum or niobium fibers were fabricated by hot pressing. Thermal

conductivity measurements showed that the fibered pellets had radial thermal conductivities two to three times higher than unfibered pellets.

Specimens were placed in the MTR in NaK capsules in order to determine their irradiation behavior. Some of the pellets were irradiated bare. Others were jacketed in Zircaloy-2 with the annulus between pellet and tube filled with either metallic lead or helium. The pellets were $\frac{3}{8}$ in. in diameter and $\frac{3}{8}$ in. long. They were irradiated to burnups ranging up to 35,000 MWD/T and at central temperatures ranging well above the melting point of the molybdenum fibers (2625°C).

Fracture and metallographic sections of irradiated specimens showed that the metal fibers tended to reduce cracking, central void formation, and the formation of large radial grains. The molybdenum fibers in the center of many of the specimens had melted and in some cases had agglomerated into a sphere of metal as large as $\frac{1}{8}$ in. in diameter. The niobium fibers did not collect into globules but reacted with the oxide fuel.

Fission gas release from unclad fibered pellets was a maximum of 15.3% of theoretical at a burnup of 32,000 MWD/T and an integral $\text{kd}\theta$ value of 81 w/cm. Gas release from similar unfibered pellets was a maximum of 6.3% at 21,000 MWD/T and an integral $\text{kd}\theta$ of 108 w/cm. The disparity of gas release is attributed to the breaking into pieces of the unfibered pellets during irradiation, dispersion of the pieces in the NaK capsule, and consequent operation at significantly lower temperatures.

3. Nondestructive Testing Techniques

a. Ultrasonic Techniques - Ultrasonic techniques are useful for measuring the properties of materials and for the nondestructive evaluation of nuclear reactor materials. The ultrasonic techniques program at the Laboratory includes Lamb wave propagation and attenuation studies of engineering materials at elevated temperatures.

Lamb wave tests were made as an experiment on the stainless steel-uranium oxide dispersion type fuel plates for BORAX-V. Four fuel plates having uranium oxide contents of 18.3 gm, 34.1 gm, 28.9 gm, and 54.0 gm were studied. Preliminary experiments were made to insure that the Lamb wave modes were being generated at an fd product such that d is the thickness of the whole plate and not just the clad. This, then, means that the Lamb wave is traveling throughout the whole plate and should be affected by different core compositions.

Measurements were made in each plate of the attenuation of the various modes. Specific attention was given to the mode the phase velocity of which equalled the longitudinal velocity, since it is the least attenuated. The distance that this mode traveled in the various plates differed by only 0.25 in. This difference is no greater than the difference obtained in successive attenuation measurements. Although the core composition affects the attenuation, such changes as do occur are of the same order of magnitude as the experimental error.

An investigation is also being made of the velocity and attenuation in three steel cylinders of Type SA-212B which are about $\frac{3}{16}$ in. in diameter and $1-\frac{7}{8}$ in. long. Because of the specimen geometry, and because the sound waves which are generated in these cylindrical wave guides will consist of a main pulse accompanied by trailing pulses, the initial pulse length must be significantly shorter than 1.3 microseconds. The Sperry Ultrasonic Attenuation Comparator does not seem to have sufficient power for these specimens; the Arenberg Oscillator is now being modified to try to produce a pulse of 0.5 microsecond duration.

b. Neutron Techniques - The photographic detection of neutron images is currently being investigated. At least one long range objective is to apply neutron radiography as a useful inspection method for nondestructive testing. Because neutrons have little effect on normal photographic emulsions, it is necessary to use intermediate converter materials next to the film. These materials emit photographically detectable radiation when bombarded with neutrons, and thereby improve the photographic speed of the detection process.

A plastic phosphor intensifier screen has been fashioned after the technique described by Stedman,* and subjected to preliminary neutron photographic tests. This phosphor, containing natural LiF, ZnS (Ag) and lucite powder seems to suffer from the same difficulty as the commercially available boron polyester scintillators used previously, namely, nonuniform efficiency of emission which results in mottled pictures. The natural lithium screens had approximately the same speed as the natural boron scintillators.

* R. Stedman, Rev. Sci. Instr., 31 1156 (Oct., 1960)

Image resolution studies have indicated that appreciable gains in sharpness can be obtained by using a single converter screen instead of double screens. This is accompanied by a loss of about a factor of two in speed. The thickness of single screen which will yield the best film density for a given neutron exposure has been determined for several metals. The best density results have been found for 0.010 in. rhodium, 0.015 in. silver and 0.020 in. indium, all by the direct exposure method.

A neutron radiographic application study has been made on the visualization of internal structure in metals. By means of a neutron radiograph it is possible to observe areas of differing neutron cross section within the metal under study. These differences in cross section are related to the crystallinity and grain structure of the sample. In general, greater neutron transmission is obtained in single crystal areas than in polycrystalline areas. For metals having interplanar spacings in the same order as the neutron wavelength, the neutron transmission through the sample will increase with grain size.

The most striking results have been obtained by the examination of nickel disks cut from a cast rod. The neutron radiographs display patterns very similar to those obtained by polishing and etching the nickel surface to make the grain structure visible. Differences are observed however, because in the case of etching only the surface structure is made visible, while the neutron radiographs display the structure throughout the thickness of the sample. The sample thickness has been varied from 0.050 in. to 0.200 in., with best results occurring for a sample thickness of about 0.100 in. Using published cross section values for polycrystalline and single crystal nickel (19.8 and 14.1 barns, respectively) to determine relative neutron transmission through the sample, good agreement with observed photographic exposures for light and dense areas on the image of the nickel has been obtained.

C. Reactor Materials Development

1. Irradiation Damage in Steels

a. Magnetic Properties - Exploratory inductance bridge measurements were performed in the shielded caves with irradiated magnet bars prepared from the remnants of multi-notch SA-212B impact specimens. About half of these bars were deformed enough to prevent their insertion into the bores of the solenoid search coils. New search coils with larger bores are being wound for the inductance bridge. Generalizations are not possible because unrelated heat-treating cycles are involved. Similar measurements on irradiated Type 304 stainless steel magnet bars gave indications of changes of magnetic properties. As in the case for the SA-212B magnet bars, the austenitic stainless steel specimens were also deformed in the impact resistance determinations, preventing complete insertion of the bars within the solenoid bores.

Inductance bridge measurements of the magnetic properties of SA-212B standard magnet bars revealed that the longitudinal and transverse bars (referring to the final rolling direction of the plate from which they were prepared) with equivalent heat treatments, were indistinguishable from each other. Measurements also showed that the as-quenched hardness of the SA-212B material prepared at the Laboratory was higher than that of the earlier experimental bars prepared by torch-heated specimens. Although the hardness falls progressively from a maximum of $R_C 48/50$ (as-quenched condition) as a function of the tempering temperature, the magnetic properties of the tempered bars were almost identical. Bars were tempered individually one hour at 700°F, 800°F, 900°F, 1000°F, and 1150°F.

The inductance bridge measurements at 100 cps frequency were confirmed by 60 cps examinations on a Magnaflux FS-300 Magnetic Materials Comparator designed for sorting of mixed ferromagnetic parts. Additional bar standards are being prepared for the study of the influence of tempering temperatures on magnetic properties below 700°F and above 1150°F, because the steel tempered at the highest temperature (1150°F) is still magnetically harder than fully annealed material (1450°F-1475°F). A group of standards was returned for reannealing at the same temperatures for additional periods to determine the effect of time at temperature.

b. Pressure Vessel Steel Irradiation - Approximately four hundred $3\frac{3}{4}$ in. long SA-212B miniature samples in the form of tensile specimens, round multi-notch impact bars, round magnet bars, and rectangular hardness bars were received for loading into helium-filled capsules. These capsules will be fitted and loaded into the "bare" and "shielded" irradiation thimbles of the EBWR. Capsules containing foils for the measurement of neutron energies are currently being exposed in EBWR.

D. Heat Engineering

1. Axisymmetric Free-Convection Heat Transfer Study.

Knowledge of heat transfer and fluid flow is required for predicting the temperature distribution and the conditions at which failure occurs in fast reactor fuel pins in TREAT experiments. For this purpose an axisymmetric free-convection problem along a vertical thin cylinder with coolants having small Prandtl numbers is being studied. The free convection field along a vertical hot cylinder presents an axisymmetric "boundary layer" problem in which the effect of transverse curvature must also be included. The primary concern of this study is the prediction of the heat transfer coefficient.

The problem has been formulated in terms of a system of partial differential equations. Since no "similar solutions" of the problem (with the given boundary conditions) are possible, the Karman-Pohlhausen integral method was used to simplify the analysis and obtain an approximate

solution. The problem has been reduced to a solution of two simultaneous differential equations. It has been programmed for numerical calculation.

2. Hydrodynamic Instability

Tests were run with a $\frac{3}{8}$ -in. test section and a $\frac{1}{2}$ -in. riser in the Armadilla loop. These tests are currently being analyzed and compared with other data. The gamma void detection equipment is being altered to get a faster response by transferring equipment from the small loop to the Armadilla loop.

It is now possible to vary the power input to the Armadilla loop sinusoidally and to record both power and system parameters during such variation. The resultant trace will be used as a check for an analog model currently being developed.

3. Measurement of Power-to-Void Transfer Function

The series of measurements in the small scale loop on a rectangular test section has been completed. Transfer functions between heating power and void fraction at different heights along the section have been measured at 27, 41, 54, and 68 atm. Inlet velocities have been varied from 80 to 160 cm/sec, and the heating power from 48 to 112 kw/l.

Nodes in the amplitude vs frequency curve similar to the results reported by Zivi and Wright* at atmospheric pressure were found in all cases. However, this node falls at frequencies far below the points predicted by earlier theories. Effects that may explain this discrepancy have been found and the theory is now being revised.

4. Void and Velocity Distributions in Two-Phase Flow

A technique is being developed which can be used to obtain the kinetic energy distribution of a two-phase stream. A pitot tube has been used, but the values obtained are much too large. The source of error is still in doubt.

The probe used for void measurements was used to count the bubbles passing a point in the stream. This was done by putting an electronic counter in the probe circuit. Whenever a bubble contacted the probe tip, the circuit was broken and the bubble registered. Results show a very high bubble frequency near the wall, and a low frequency at the center. If the void distributions are also used, this data would indicate that bubbles near the wall are very small ($\theta_B \approx 0.01$ sec) and at the center much larger ($\theta_B \approx 1$ sec). θ_B is the time length of the bubble. Accurate values of the velocity distribution will allow a space length of the bubbles to be calculated.

* S. Zivi and R. W. Wright, "Power-Void Transfer Function Measurements in a Simulated SPERT-I Moderator Coolant Channel," ANL-6205.

5. Packed Bed Reactor Studies

The problem of shutdown cooling for a packed bed experiment in CP-5 was continued. An analytical expression for the decay heat generation rate was applied with previously completed analyses of the thermal characteristics of beds of interest, leading to the conclusion that forced convection cooling will be required for about three hours after shutdown from a short term irradiation. The conservative, one-dimensional analysis indicates that at the end of this period heat dissipation by radiation and conduction alone should limit peak bed temperatures to about 760°C in the CP-5 test hole and to about 600°C with the experiment removed from the thimble. These cooling requirements should be met to avoid confusing effects which may have occurred during irradiation with forced convection and high temperatures.

6. Boiling Liquid Metal Experiment

A small scale loop designed for measurement of vapor volume fractions and two-phase frictional behavior of flowing sodium is under construction. Materials and instrumentation have been ordered and partial delivery obtained.

The low temperature NaK-Argon loop is complete and will be operated to test the feasibility of measuring void fractions with an electromagnetic flowmeter.

A test has been initiated to reflect the stability and calibration errors of the proposed inductive liquid level gage in the boiling loop temperature range from 220°F to 1700°F .

7. Physical Behavior of Reactors and Reactor Systems

Analytical phases of the study of the effect of the heat flux distribution on the Nusselt number for liquid metals flowing between parallel plates, assuming a constant velocity and neglecting convective heat transfer, has been completed. It delineates the dependence of the coefficient of heat transfer, η , on the shape of the heat flux distribution. It also permits an estimation of the error encountered in using experimental values of η for uniform heat flux distribution to make calculations in the case of non-uniform distributions. In addition, the numerical work required to determine the distribution influence has been outlined.

Further attempts have been made at solving the equations for liquid metal coolants in single-phase flow between parallel flat plates. No improvement in obtaining tractable forms has been realized although some advanced forms for solutions when slug flow is assumed have been obtained.

8. Dynamic Correlation Measurements of Heat Transfer and Fluid Flow Characteristics

Computing Code RE-245 was used to compute the power density spectrum of the steam void fraction for two sets of data, each consisting of 1000 data points. The results have a large scatter, and even negative spectral density values are obtained. This is mainly due to the low signal-to-noise ratio inherent in the dynamic void measurements by the gamma-ray technique. Longer records would also decrease the scatter. However, the results seem to indicate that the power density spectrum of the steam void fraction is a white noise in the analyzed cases.

E. Separations Processes

1. Fluidization and Fluoride Volatility Processes

a. Direct Fluorination of Uranium Dioxide Fuel - Pilot plant studies of a direct fluorination process for the recovery of uranium dioxide fuel are being continued with emphasis on complete fluorination of hydrogen-fired pellets. Temperature control was successfully maintained within a broad range for operational purposes at an average temperature of 500°C by a varying fluorine flow rate at constant nitrogen flowrate (average fluorine concentration 45 percent). The total flow rate was about 0.4 ft/sec. The average bed temperature of 500°C was maintained over a wide range of heat transfer conditions in three different runs in the three-inch diameter air-cooled reactor with six-inch deep beds of $\frac{1}{2}$ -inch diameter pellets. In one run the fluorination was performed in 9.5 hours without an inert bed to aid heat removal from the reaction zone. Large temperature differentials existed in the reaction zone from the control temperature of 500°C to the wall and skin temperatures of 50°C. A run was made with calcium fluoride powder as an inert fluidizing medium to provide more effective heat removal from the reaction zone. Oxygen rather than nitrogen was used as the diluent gas to test its effect on reaction rate. The gas flow was not sufficient to maintain fluidization. Highly nonuniform pellet bed temperatures were obtained with the same 50°C wall temperature, and the control temperature fluctuated $\pm 50^\circ\text{C}$ from 500°C; however, the fluorination was successfully carried to completion in less than 9 hours. No difficulties in operational behavior could be attributed to oxygen. In another run using nitrogen as a diluent, the superficial gas rate was increased from about 0.4 to 0.61 ft/sec (at process conditions of 45 percent chlorine and 500°C bed temperature); this condition evidently prevented caking of the fluid bed, and excellent heat transfer was demonstrated in the fluorination (wall temperature about 480°C). In this run the pellets were 80 percent reacted in 11.5 hr. Overall production rates in these runs ranged from about 15 to 30 lb $\text{UF}_6/(\text{hr})(\text{sq ft reactor cross sectional area})$.

b. Processing Stainless Steel-Clad Fuel Elements - A two-zone fluid-bed reactor is being used to study chlorination and fluorination reactions for the decladding and dissolution of stainless steel-clad uranium dioxide fuel elements. The chlorination products (iron and uranium chlorides) formed in the lower zone pass into the upper zone where they are converted to the corresponding nonvolatile fluorides by reaction with hydrogen fluoride. The reaction of stainless steel closed-end tubes with 48 mole percent chlorine in nitrogen at 625°C (as indicated by weight loss) decreased slightly with time; however, the time dependence was not so great as at a lower temperature (575°C). Complete reaction of the specimen was achieved in 3.8 hours under these conditions. Data from several runs showed that the penetration rate dropped significantly from 11 mils/hr at 48 percent chlorine in nitrogen to 4 mils/hr at 27 percent chlorine concentration.

High bed temperatures may be achieved without subjecting the reactor walls to extreme temperature conditions by means of an internal heat source such as an exothermic reaction. In a separate fluid-bed experiment at 625°C where the exothermic hydrogen-chlorine reaction was tried, a penetration rate of only 4 mils/hr was observed although the exothermic reaction successfully maintained the high bed temperature. The reduced rate was attributed to the fact that excessive chlorine was consumed by the hydrogen, leaving a relatively low concentration to react with the stainless steel 304 specimen. The hydrogen chloride formed by the auxiliary reaction is considered an inert in this case since its reaction rate with stainless steel is about 100 times lower than that of chlorine. This experiment indicates that the use of other exothermic reactions might prove feasible.

In an additional experiment, an attempt was made to ignite stainless steel with chlorine using lower concentrations than those used previously. When 27 mole percent chlorine in nitrogen was used, only sintering (no ignition) of the nonvolatile chlorides and the bed material occurred on the tube specimen at temperatures up to 670°C. In an earlier experiment with 87 mole percent chlorine in nitrogen, the stainless steel ignited at 645°C.

In order to determine whether unusual corrosion rates of stainless steel might be achieved in dissociated chlorine, preliminary experiments were made in a beta-gamma radiation field (8.3×10^6 rad over a period of 349 minutes) and in the discharge region of a Tesla coil. No unusual reaction rate effects were produced.

c. Plutonium Fluoride Studies - An important problem in the Direct Fluorination Volatility process is the removal of plutonium as plutonium hexafluoride from a solid substrate by the reaction of fluorine with plutonium oxide. Experiments are being performed to study the effect of time and temperature on the removal of plutonium from various solid substrates such as zirconium tetrafluoride, calcium fluoride, fused alumina and silica.

The decomposition of plutonium hexafluoride gas has been studied further. After an initial fluorination with a small amount of plutonium hexafluoride, the rate of decomposition in a 53-ml nickel vessel at 80°C with an initial starting pressure of approximately 100 cm of plutonium hexafluoride was about 1.5 percent per day.

2. General Chemistry and Chemical Engineering

The attainment of lower cost nuclear power depends on the development of more economical methods of manufacture of nuclear fuel materials. Research and development for devising new chemical methods or improving chemical engineering techniques or equipment is being conducted.

The fluid-bed conversion of uranium hexafluoride into uranium dioxide is being studied. This is an essential step in the production of enriched oxide for nuclear reactor fuels. The process involves either a single-step pyrohydrolysis and reduction using steam and hydrogen simultaneously, or a two-step process using alternatively steam or hydrogen, followed by the other reactant.

Development of a small diameter fluid-bed calciner for the direct conversion of plutonium nitrate to plutonium dioxide has begun.

Conditions for liquid metal reduction of the oxides of uranium, plutonium, and thorium are being explored. Preparation of refractory uranium and plutonium compounds is being investigated.

a. Steam Hydrolysis of UF_6 to UO_2F_2 - The excessive formation of fines has been a major problem in preventing extended operation of the three-inch fluid-bed reactor for carrying out the hydrolysis of uranium hexafluoride. Attempts are being made to alleviate the problem. Changing the location of the product take-off to the top of the reactor instead of the bottom which is in the vicinity of the hexafluoride inlet point and lowering the gas velocity (0.375 ft/sec instead of 0.75 ft/sec) did not relieve the fines forming problem. Column modifications are planned which will allow entrainment and separation of the fines from the bed.

b. Reduction of UO_2F_2 to UO_2 - Fluid-bed studies of the reaction of uranyl fluoride with hydrogen-steam mixtures to produce uranium dioxide were continued. Results of early experiments at 650°C for 4 hours were as follows:

Steam in Steam-Hydrogen Mixture (volume %)	Residual Fluoride (weight %)
7	0.09
12	0.07
50	0.02

c. Calcination Studies in Small Diameter Columns - Work is proceeding on a dual purpose scheme for direct conversion of plutonium nitrate to plutonium dioxide having as aims: (1) to demonstrate that this may be accomplished in small diameter columns (6-inches or less in diameter), and (2) to reduce the total volume of off-gas which is normally encountered in a fluidized-bed calciner using air as the fluidizing gas. By placing the liquid feed atomizing nozzle in the bottom of the unit, directed upward, the atomizing air plus the evaporated feed and gaseous decomposition products are used as the fluidizing medium, thus dispensing with fluidizing air. Several successful short (about 1 hr) runs were made in a $2\frac{1}{4}$ -inch stainless steel column. Work will continue in this column to optimize conditions.

d. Preparation of Uranium and Plutonium Compounds - The possibility of preparing various refractory uranium and plutonium compounds in liquid metal solutions was further explored. Uranium monocarbide has been precipitated from both cadmium and zinc systems. A single compound, either USi_2 or USi_3 , was prepared by precipitation of uranium with silicon from a cadmium solution. Preparations are being made to prepare plutonium monocarbide in liquid magnesium and liquid cadmium solutions.

3. Chemical - Metallurgical Process Studies

a. Calorimetry - Studies of the combustion of uranium in fluorine to form uranium hexafluoride are being continued. Emphasis is being placed on reducing the amount of lower fluorides that are formed during the combustion.

Calorimetric combustions of cadmium and hafnium are being carried out.

Preliminary experiments to develop techniques for burning tantalum and niobium in fluorine have been completed. Preliminary studies of the combustion of vanadium in fluorine and of zirconium hydride in oxygen have been started.

The value for the standard heat of formation of boron trifluoride was revised from -269.93 kcal/mole to -270.16 kcal/mole. The revision was based on additional information that has been obtained on the reaction of nickel bomb fittings during combustion of boron and boron nitride in fluorine.

b. Liquid Metal Solvent Studies - Crystals obtained from a zinc-uranium melt held at 800°C were found to be the epsilon uranium-zinc intermetallic compound. The uranium content of the crystals was determined to be 25.9 weight percent. This represents the best available information concerning the composition of the epsilon phase just below its peritectic decomposition temperature (about 840°C). The zinc content of the intermetallic compound has not yet been determined.

An electrolytic method has been developed for an analytical separation of rare earth-cadmium intermetallic compound crystals from excess cadmium. The cadmium matrix is dissolved at the anode in an aqueous solution that contains 120 g of sodium cyanide and 22.5 g of cadmium oxide per liter. A nickel cathode is used. The electrolysis is carried out at a current density of about 0.2 amp/sq cm. As the electrolysis proceeds, the crystals of intermetallic compound, freed of the cadmium matrix, fall to the bottom of the beaker.

Decomposition pressures of UCd_{11} have been determined by the effusion method over the temperature range of 305° to 380°C . The relationship between the equilibrium pressure and the temperature ($^\circ\text{K}$) may be represented by the empirical equation.

$$\log P_{\text{mm}} = 9.060 - \frac{6105}{T}$$

Final data on the magnetic susceptibility of the uranium-cadmium intermetallic compound have been obtained. The susceptibility values follow a Curie-Weiss relation ($\chi = C/T - \Delta$) with $C = 1.27_6$ and $\Delta = -30\text{ K}$. The observed effective Bohr magneton number of 3.21 indicates that there are two unpaired electrons. This value is smaller than required for j-j coupling ($\mu_{\text{eff}} = 3.58$) but too large for spin-only coupling.

F. Advanced Reactor Concepts

1. Compact High Power Density Fast Reactors

The development of a high power density fuel tube assembly for a tentative core design is the present goal. Assuming an average heat flux of 1.5 w/cm^2 , it appears that a power density of 1 Mwt/liter of core volume is readily obtained. In addition, a core design is pursued in which there are two sections: (a) a tubular section for heating the sodium coolant to saturated conditions, and (b) a woven wire fuel section for superheating the sodium.

The basic compact reactor being analyzed may be useful for the following applications:

a. Systems where ultra compactness and light weight are required. The direct cycle sodium-cooled reactor would be a likely choice.

b. Ship or other mobile devices for which binary sodium-mercury plant could be adapted.

2. Fast Reactor Test Facility (FARET)

The survey to determine the need for and potential value of a FARET facility has been completed. A list of interesting fields of investigation for which the facility would be most useful has been developed as follows:

- (1) Experiments that should permit operation for thermal burnout conditions to determine operating margins of existing or planned reactors.
- (2) Experiments in which the rate of release of fission products will be observed.
- (3) Physical changes in fuel with nuclear heating and modest burnup such as expansion, melting, swelling, etc.
- (4) Local fuel evaporation and condensation effects.
- (5) Effect of rate of heating on core and fuel structure.
- (6) Clad and fuel integrity at high temperature.
- (7) Reactivity coefficient of fuel heating. The Doppler effect and the coolant void coefficient of a small and large reactor would be different. The Doppler coefficients would be obtained from critical experiments whereas the coolant void coefficient could be eliminated as far as possible in a small core by providing for very high coolant flow rates.
- (8) Boiling liquid metal reactor studies, particularly the dynamics of the systems.
- (9) Fuse tests for fast-thermal coupled systems. These could be of interest for a modest size facility. Calculations have indicated that the percent reactivity in the thermal section and the prompt neutron lifetime do not change greatly in going from a small to a large core.
- (10) Final proof testing of a core.
- (11) The determination of the dynamic behavior of the system for physics at various conditions of flow and mechanical-thermal parameters. These could possibly be done with oscillator tests, but limited excursions might give more direct information about the power coefficient of reactivity of the fuel. Such excursions would probably be permissible in this system while they would not be in any other.
- (12) Development of coolant purification techniques under actual reactor operating conditions.

(13) Reliability testing of complete systems.

(14) Fuel development. It should be possible to incorporate fuel sections containing different experimental fuel of advanced nature.

(15) Doppler coefficient tests by degrading of the neutron spectrum by small amounts of fission moderator.

3. Direct Conversion Survey

Effort is directed toward a brief study of the application of nuclear fission energy for direct energy conversion schemes. Particular emphasis is placed on the fuel cell type systems which employ reactants that can be regenerated in situ or remotely in a nuclear reactor.

Table VI is a summary of some of the parameters used for comparison of the various direct energy conversion schemes.

Table VI. DIRECT CONVERSION SCHEMES POTENTIAL

System	Current Size Range	Weight/Power Ratio (kg/kw)		Efficiency (%)		Energy Weight Ratio (watt-hr/kg)
		Present	Projected	Present	Projected	
Thermoelectric (Lead telluride)	Up to 5000 watts	60	20	5	15	----
Thermionic Conversion	Several hundred watts	30	10	10	30	----
Photovoltaic	Up to several hundred watts	1000	--	10	20	6600
Magnetohydrodynamic	12 kw	----	--	50% as a topping unit for steam cycle		----
Fuel Cells						
H ₂ + O ₂	Several kw	30-15	--	70	--	
Regenerative LiH	Several hundred watts	100	--	40	--	
Petroleum Gas (Allis-Chalmers)	15 kw	61	--	--	--	

For purpose of reference, an automobile engine has a weight-to-power ratio of approximately 4 kg/kw and an efficiency of 15 to 20%. For rotating turbine generators this ratio is approximately 1 kg/kw.

Table VII has been prepared to show the range of weight-to-power ratios for various heat sources.

Table VII. Heat Sources for Direct Conversion Schemes

Heat Source	Weight/Power Ratio, kg/kwt	
	Without Shield	With Shield
Isotope low specific activity		
Sr ⁹⁰	11	---
Ce ¹⁴⁴	7	
Isotope high specific activity		
Po ²¹⁰	0.2	---
Cm ²⁴²	0.1	---
Reactor		
50 kwt	2.5	25
500 kwt	0.45	3.5
5000 kwt	0.30	1.0

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