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REACTOR DEVELOPMENT PROGRAM PROGRESS REPORT

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DISCLAIMER

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 report issued in this series are:

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I. WATER COOLED REACTORS

A. EBWR

1. Preparation for High Power Operation

a. Reboiler Building Equipment Checkout - The turbine-generator set, one of the two reboilers, and the air-cooled condenser systems were tested with reactor steam. Approximately 90,000 lbs of steam per hour were fed to one reboiler to produce 350 psig secondary steam. This steam condensed in the air-cooled steam condensers and returned to the reboiler. This rate of operation is equivalent to 30 Mw of thermal heat and is about 75% of the load that each reboiler was designed to handle. A maximum radiation level of 270 mr/hr at 2 in. was measured on the pipe line between the reboiler and its drain tank during this test.

A number of minor mechanical problems were uncovered during this test and are being corrected.

b. Turbine Governor Leaf Spring Failure - On July 10, 1962, the EBWR reactor was operating at 20 Mwt producing 4 Mwe energy when the reactor was scrammed due to the failure of a leaf spring in the turbine governor.

The failure was attributed to the flow of static electricity from the turbine rotor through the steel and bronze gears in the governor. The flow of current resulted in the formation of pits in the steel driving gear causing excessive wear on the bronze driven gear. Whereas the backlash between the gear was originally 0.006-0.008 in., it had increased to 0.018 in. after 428 hr of operation.

Ground brushes were installed between the generator and exciter shaft to eliminate the current flow between the gears.

c. EBWR Transfer Functions - The transfer function of the EBWR was measured at 40 Mw and 600 psig with feedwater injected through the upper feedwater ring. The boric acid concentration was less than 0.15 gm/gal. Control rods Nos. 1 through 8 were at 28.5 in. and the center rod (No. 9) was at a mean position of 21.5 in. with equilibrium xenon. The measurements were made using the rod oscillator at 32 frequencies from 0.0012 to 9 cycles/sec. The null analyzer was not used for this experiment. Data was recorded with the analog sine-cosine cross-correlator and digital data system. The

transient response and noise were recorded. The RMS (root mean square value) of the reactor noise was measured using an analog computer.

The final experiment in this test series used discrete interval binary noise to simulate a statistical reactivity input. The binary noise was generated by using a white noise generator, square wave generator, and a custom designed logic unit. This synthesized noise signal was used to control the position of the center rod (up or down) about its mean position of 21.5 in. The first binary noise record was made with: the rod velocity set at 4 cycles/sec under free running conditions, plus or minus 1-in. stroke, 4 cycles/sec square wave period (0.25-sec binary noise interval), 0.096-sec digital sampling rate, and 864-sec record length. The free running rod velocity sets the capability of moving the rod from down to up in one-half cycle or 0.125 sec when programmed for start-stop operation. The second binary noise record was made with: the rod velocity set at 1 cycle/sec free running, ±2in. stroke, 0.5 cycle/sec square wave period (2-sec binary noise interval), 0.768-sec digital sampling rate, and 115-min record length. The following information was recorded: control rod position, neutron flux, reactor pressure, inlet water subcooling, inlet velocity, and exit velocity of instrumented fuel assembly. The neutron flux to control rod position data was cross-correlated and the power density spectrum was calculated. The record with a 2-sec binary noise interval provided information on the reactor transfer function from 0.0013 to 0.65 cps. The record with a 0.25-sec binary noise interval covered the transfer function from 0.01 to 3.2 cps. The 0.096-sec sampling rate provided resolution up to 5.2 cps, but the 4-cps control rod velocity resulted in the 3.2-cps frequency limitation in the reactor transfer function. In order to obtain reactor transfer function information at frequencies greater than 3.2 cps, it will be necessary to increase the free running rod velocity to greater than 4 cps. The amplitude and phase information obtained from the binary noise data compared very well with the data obtained with the oscillator experiments.

Analysis of the 40-Mw data and extrapolation indicate reactor stability to 85 ± 5 Mw. The extrapolation from 40 to 85 Mw is linear and represents a factor of 2.1 in power increase. System nonlinearities can be expected to alter the 85-Mw estimate. Additional data will be taken at powers greater than 40 Mw and used to determine a new stability extrapolation.

The root mean square (rms) noise measured at 40 Mw, 600 psig was 3.22%. The relation of rms noise to power and pressure is indicated in Table I.

Table I. Relation of Root Mean Square Noise to Power and Pressure

Power (Mw)	Pressure (psig)	RMS Noise (%) Without Boric Acid	RMS Noise (%) With Boric Acid
10	300	0.97	0.90
20	300	3.17	1.84
10	600	0.79	0.78
20	600	1.96	1.06
40	600	3.22	

d. EBWR Instrumented Fuel Assemblies - Hydrodynamic performance measurements were made utilizing the instruments installed in instrumented fuel assemblies II and III. Measurements of the inlet and exit liquid velocities, subcooling, and cadmium ratios were made during reactor operation at 5, 10, 15, 20, 25, 30, 35, and 40 Mw. The system operating pressure was 600 psig (41.8 atmospheres) and the reactor water level was 720 ft 10 in. Most of the data taken has been reduced and performance curves have been plotted.

e. Reactor Fluid Control System - The air booster pump has been received and will be connected to a large volume air storage tank. The ALPR test pressure vessel has been reclaimed from storage to serve as the air storage tank. The booster pump will take 80 psig laboratory air and store it in the tank at 500 psi. Air stored in the tank will be used as the supply for the flow system.

B. BORAX-V

1. Operations and Experiments

The reloading of boiling fuel assemblies was completed. It was necessary to increase the number of boron-stainless steel poison rods from 32 to 34 to maintain adequate shutdown margin and assure that the reactor is subcritical with any single control rod withdrawn. This poison addition is probably needed because of the small difference in construction between the new and the old control rods. The new reference poison rod arrangement is shown in Figure 1.

Additional flux wire irradiations and Cd-ratio measurements are being made to confirm previous data and to obtain new data. Detailed distribution of thermal flux around a control rod was measured for comparison

with results obtained through calculations previously performed with the DSN and PDQ codes.

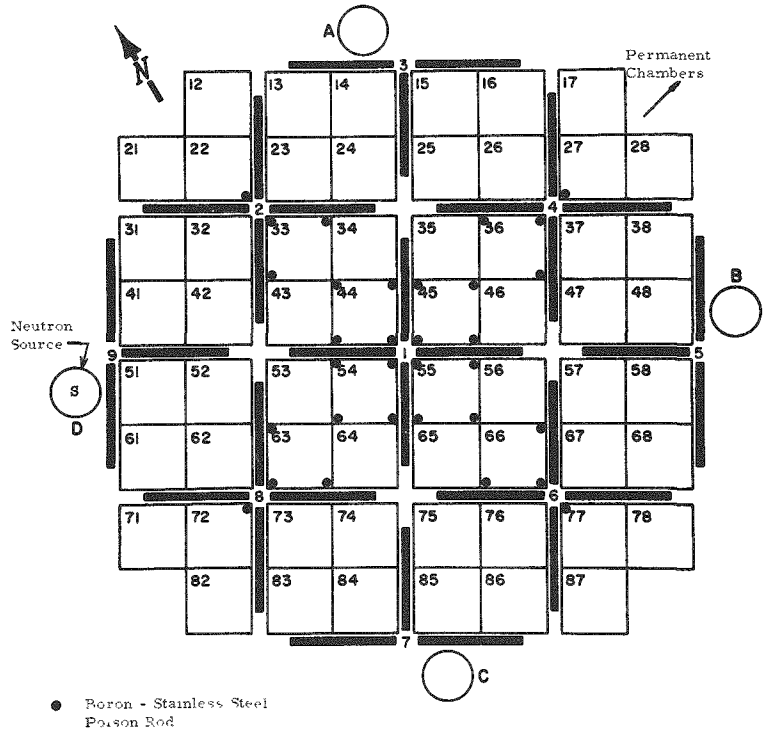

Figure 1. Final Poison Rod Location - Reference Boiling Core - BORAX-V

The oscillator has been installed in the reactor vessel, and testing to determine its reactivity worth and wave shape in the three possible locations has been started in the cold boiling core containing boric acid.

A study was made of the temperature coefficient of the BORAX-V loading, seeking an explanation of why such a low value should be exhibited during the cold critical measurements. An array of BORAX fuel was assumed. No structural material was included, although an equivalent amount of boric acid (18.751 gm/gal) was assumed. Disadvantage factors were evaluated for the boric acid condition, and constants were then obtained via the MUFT and SOFOCATE codes. Four-group, fundamental-mode calculations gave values for k_{eff} as follows:

Water Temperature	H_2O Density gm/cm^3	k_{eff}
79°F	0.99678	1.1563
112°F	0.99678*	1.1562
112°F	0.99066	1.1544

*This density is the same as the 79°F-value to check effect of change in neutron temperature.

These results indicate that a temperature coefficient, with boric acid, of ~-0.0045% $(\Delta k/k)/°F$, is due almost entirely to a change in moderator density over the region of calculation. This compares to a measured value, with boric acid, of ~-0.0002% $(\Delta k/k)/°F$.

2. Modification and Maintenance

During installation of the new control rods, a bad scratch due to a particle in the seal was found on a control rod extension shaft. As a result, all of the reactor vessel internals and control rod drive seals were removed and thoroughly cleaned. The control rods, seals, and drives were all re-installed temporarily, using two old control rod extension shafts. One of the control rod drive motors burned out and was replaced with a spare. After these changes, the control rods and drives were satisfactorily tested.

The two new control rod shafts that were sent back for repair have each been stripped and are being replated. In addition to the shafts being replated, four new spare shafts are being fabricated. These shafts are now being ground for the second heat treatment at 1100°F.

The new electric cartridge heaters were received; and the heater flanges, with 6 dry wells each, were completed. These replacement preheaters were installed, hydrostatically tested, and are now ready for operation.

New reactor demineralizer bypass piping was installed, hydrostatically tested, and placed in service. Five cubic feet of anion resin were added to the makeup water demineralizer, but definite evidence of changed performance is lacking, due to low demand for water this month.

In an effort to improve the reliability of counting data obtained from flux wires by seeking a lower radiation background, the counting facility was moved from the EBR-I Building to a trailer. A motor-generator set is being used to provide isolation from power line transients.

Range-change bellows for 3 downcomer differential-pressure cells were delivered after a delay of some 6 months and have now been installed. The new bellows will allow a minimum full-scale range of 12 in. water and a maximum full-scale range of 60 in. water, compared with 20 and 100 in. water for the original bellows supplied erroneously.

3. Critical Experiments

For discussion of critical experiments with BORAX-V fuel see III.A.3 (General Reactor Technology, Applied Reactor Physics, Thermal Critical Experiments) Hi-C and BORAX-V Superheat.

4. Procurement and Fabrication

a. Superheat Core - The examination of all fuel plates received from AI has been completed. A summary of inspection results is given in Table II.

Table II. Summary of Inspection BORAX-V Superheater Fuel Plates

Plate Type	No. Required by Contract	No. Received at ANL	No. Accepted by ANL	No. Rejected by ANL
HCD	40	40	40	0
FCD	40	40	40	0
HPD	40	42	42	0
FPD	40	40	40	0
HCE	190	192	190	2
FCE	190	190	185	5
HPE	230	230	227	3
FPE	230	230	212	18

One rejected plate, FPE 132, is being held at ANL for destructive examination to verify the results of nondestructive testing for bond integrity. The flaw which was detected by the ultrasonic through transmission test is also discernible on a radiograph of the plate as a lighter (more dense), area. The other 27 rejected plates have been returned to AI.

The enriched fuel elements required for the BORAX-V superheat loading are as follows:

Element Type	No. of Elements	No. of Subassemblies	No. of Plates
Central	13	65 (5/element)	260 (4/subassembly)
Central Instrumented	3	15	60
Peripheral	17	85	340
Peripheral Instrumented	2	10	40

The 13 required central fuel assemblies have been completed. A total of 71 central subassemblies were brazed. Of these, 10 exhibited excessive leakage of argon gas from between side strips and plates or spacer wires and plates during water immersion leak checking. Since 65 subassemblies were needed to complete 13 elements, four subassemblies were

successfully rebrazed. The six remaining subassemblies will not be processed further unless the need arises.

Processing of peripheral type plates has begun and ten subassemblies have been brazed to date.

b. Experimental Components - The oscillator drive has been modified slightly to accommodate a photocell pickup to measure accurately a preset number of rotations. Modifications have also been made to the oscillator housing and holding brackets to facilitate operation during the cold and hot criticals. The synchronous motor has been mounted on the special rotating seal. The high-temperature source drive has been modified to improve reliability.

Development work on a Zircaloy-2-to-stainless steel pressure-tight joint for the step-function generator is continuing. The second joint fabricated, using pure silver as a braze material, failed the autoclave corrosion test. A mechanical joint, using a silver-plated, stainless steel O-ring, and another, with a pure gold braze, will now be tried. The poison section of the step-function generator satisfactorily passed the autoclave test and has been shipped to Idaho.

In order to evaluate possible substitute control materials for use in BORAX-V, the following $3\frac{1}{2}$ x 8-in. sample plates are being fabricated or procured:

1 piece - Present reference design; $\frac{3}{16}$-in.-thick, 50 w/o Boral, canned and seal-welded in $\frac{1}{16}$-in.-thick, Type 304 stainless steel

5 pieces - Hafnium; $\frac{1}{16}$-in. thick

5 pieces - 2 w/o natural B, Type 304 stainless steel, $\frac{1}{16}$-in. thick

5 pieces - 2 w/o, 93% enriched B^{10}, Type 304 stainless steel, $\frac{1}{16}$-in. thick

The relative reactivity worths of these samples will be compared in BORAX-V.

Equipment for an analog computer transfer function measuring system similar to that used on EBWR has been ordered and will be used in conjunction with the null balance method. In an attempt to minimize hot spots in adjacent fuel rods, aluminum filler pieces have been placed at the corners of the dummy fuel assembly which houses the oscillator rod to remove the water and cut down flux peaking.

The first instrumented boiling fuel assembly (II) was delivered to Idaho. This unit utilizes the modified pressure seal design with Conax

packing glands. Silver brazed junctions in its terminal box have been completed, and the assembly is now installed in the air-water test loop for flowmeter calibrations.

Salvage work on the damaged pressure seal block of instrumented assembly I is planned to be carried out by splicing on leads to those damaged, and welding the splicing sleeves into a modified seal block. Splicing sleeves would be induction-brazed to the thermocouple sheaths.

Vacuum-brazing of a dummy, instrumented superheater fuel element, using Coast Metals 60 alloy, resulted in void spaces at the fuel plate thermocouple tips. Modification of the brazing process to eliminate the voids is under investigation.

5. Development and Testing

a. Corrosion Tests - The boric acid (5.28 g/l) corrosion test on an X8001 aluminum-Type 304 stainless steel couple at 250°C and 600 psig was terminated after 816 hours. Neither alloy showed any evidence of severe corrosion. There was no pitting of the aluminum where it touched the stainless steel. Weight change measurements indicate that the corrosion rate of the aluminum was about 2 $mg/dm^2/day$. The stainless steel had only a very thin gray-brown adherent film. This work indicates no troublesome corrosion should be expected in BORAX-V during high-temperature runs with boric acid.

b. Corrosion of Structural Materials in Superheated Steam - A contractor has started the installation of the major components of a steam dynamic test facility. Ultimately, tests at 200 ft/sec, 650°C, 1000 psi will be possible. Exploratory tests in small refreshed autoclave systems are continuing until the loop is complete which is estimated to be October 1962.

Sample surface preparation plays an important role in determining the total amount of corrosion of the 300 series stainless steels at 650°C. This is particularly true in the presence of oxygen. Electropolished specimens prepared from baths of two different compositions suffer much more corrosion than wet ground specimens. Etched samples are intermediate. The amount of metal loss can be quite high. For example, about 0.001 in. of SS-347 was lost in one week for an electropolished specimen exposed to steam containing 0.1-0.7 ppm oxygen. No information has been obtained concerning the shape of the metal loss corrosion curve. It is believed that the corrosion rate decreases sharply during the first week, as judged from the amount of liberated corrosion product hydrogen.

c. Recombiner Tests - The H_2-O_2 recombiner test rig was ready for operation when a detonation occurred in the Knall-gas generator, destroying a pressure gauge and flow rator. The cause of ignition is still

unknown. A new method of injecting hydrogen and oxygen into the boiler separately, which should reduce any further chance of an explosion, is being considered.

d. Continuous Chloride Analyzer - The automatic chloride analyzer has been checked out and operates satisfactorily when ambient temperature is controlled to ±5°C at around 25°C. The instrument can detect 0.05 ppm chloride with an accuracy of about ±50% and is sensitive to 0.01 ppm with unknown accuracy.

e. Advanced Superheater Fuel - The six developmental cermet fuel plates made of depleted UO_2 dispersed in a matrix of Type 406 stainless steel and clad with Type 406 stainless steel, which were fabricated by Atomics International, were destructively tested by the supplier. An ANL inspector found these plates to be satisfactory. Negotiations are in progress with AI for an addition to the original superheater fuel fabrication contract covering the fabrication of 50 Type 406 stainless steel plates. These plates will be assembled into two experimental superheater fuel assemblies by ANL and tested in BORAX-V.

Development work on vacuum brazing of Type 406 fuel plates into four-plate elements has started. Thus far, sample brazes using Coast Metals 60 alloy at 1170°C and Coast Metals 608 and 57 Special alloys at 1000°C have been tried. Only Coast Metals 60 produced a bond, but the braze alloy did not flow evenly and left unbonded gaps. In contrast to the bright appearance of Type 304 stainless steel when removed from the furnace, the Type 406 stainless steel has a dark oxide coating. The 1170°C brazing temperature caused the growth of large-size grains in the Type 406 alloy. Below 980°C, this grain growth does not occur. A sample braze using plated Kanigen-nickel as the braze material is being prepared.

II. LIQUID METAL COOLED REACTORS

A. General Research and Development

1. ZPR-III

Work continued on Assembly 40, an 8-region cylindrical reactor which contains a partial-density axial beryllium reflector and a full-density radial beryllium reflector. The axial reflector is adjacent to the core, while the radial reflector begins 9 cm from the core edge. The dimensions and volume fractions were given in (ANL-6580 Progress Report, June, 1962).

Initially, the reactor was loaded and brought to criticality with no beryllium in the axial reflectors. It was then determined that normal reactor operations would be safe with beryllium in the axial reflector. A single core drawer containing beryllium in the axial reflector region could be removed from the reactor and the control rods containing fuel could be moved through the beryllium with no net positive reactivity effects. The axial reflector regions were then loaded with beryllium.

The experimental program following the complete loading of the reactor has included control rod calibration, worth of fuel at the radial core edge, reactivity coefficient measurements, axial and radial fission traverses, and central fission ratios. Reactivity coefficient measurements were made for a large number of samples of both fissile and nonfissile materials at three locations - the core center, radial core edge, and axial core edge.

2. ZPR-VI and ZPR-IX

a. Building - The construction of the emergency venting system for the reactor cells has been completed, and the installation of the control circuitry is underway. The revised calculations of moments, stresses, and strains for the containment cell were completed (ANL-6525, Progress Report for February, 1962). These calculations were made at 44 strain gauge locations for internal pressures of 2, 4, 6, 8, and 10 psig. These are now being compared with the measured strains.

b. ZPR-VI Assembly - Preoperational testing is continuing. The matrix tube bundles were realigned and their location accurately determined in preparation for the measurement of deflection under loaded conditions. Thirty metric tons of depleted uranium blocks will be used to perform the sag test. Both the movable and stationary halves will be tested in turn.

The performance tests conducted on the danger coefficient sample changer and analog table position indicator indicated the need for some minor modifications. A new gear train has been installed in the drive

mechanisms of the former, and the corrective machine work on the latter is nearing completion. Detailed drawings for a conveyor system to move loaded fuel drawers are being prepared.

3. AFSR

The canned lower section of AFSR fuel was returned to Idaho, quarterly maintenance was completed, and the lower core section installed. The reactor was brought to criticality in three runs by stepwise insertion of the shim rods. The final value for fuel loading is 21.665 kg of enriched uranium. This slight discrepancy from the value previously reported (21.667) is due to minor machining of one of the uranium discs prior to canning of the lower core section.

Total excess reactivity for the final configuration of the core with all experimental holes empty is approximately 250 ih. The increase over the preliminary value obtained last month with the fuel disc in an unsealed can is apparently due to a more snug fit of the welded can, permitting a closer assembly of the core. The position indicator for the lower core section supports this explanation.

Since returning to regular operation, AFSR has made a number of runs to:

1. irradiate scandium oxide and gold samples for the spectral activation program,
2. continue testing of the new model period meter,
3. check an experimental lithium-loaded counter for ZPR-III,
4. supply activated samples for checking BORAX-V counting gear, and
5. continue experiments with the equipment for fluctuation analysis.

It appears that much of the trouble with the equipment for fluctuation analysis has been remedied by improving the ventilation of the electronic systems.

Machine shop work is continuing on the criticality monitor and the electronics is essentially complete. First in-pile tests are scheduled during August. Photomultipliers and filters are on hand for investigating the color response of various scintillators to gammas and neutrons.

B. EBR-I

1. Mark III Operation

A number of irradiations were performed in the course of operation for Laboratory Divisions and for Phillips Petroleum AED.

In connection with studies of irradiation-induced defects and consequent effects on mechanical properties, two capsules containing cobalt oxide and magnesium oxide single crystals received 1 and 35 hours of irradiation time, respectively, at low temperature, full power.

Two capsules were irradiated for MTR personnel as part of a program for developing threshold detectors to measure fast neutron fluxes in test facilities and for studying n, p reactions in rare earth nuclides.

Other irradiations included materials for comparison of the decay rate of gross fission products of U^{235}, U^{238}, and Pu^{239} and for a study of the activation cross sections of unstable nuclides. Two notched impact samples with attached thermocouples were measured at equilibrium temperatures for the Reactor Engineering Division for application to an identical irradiation performed during the month of June.

During the month, a group of technicians and supervisors from the EBR-II operations organization participated in reactor operation as part of the training program. Also, in conjunction with the training program, gold foils were irradiated. Their arrangement in the reactor was devised to indicate the vertical flux pattern in the graphite portion.

Cave modification continued during the month; the exhaust fan was received, and installation started.

2. Fabrication of Core IV Components

The final lot of 0.080 in. O.D. Zircaloy-2 instrument tubing is in the process of being evaluated. To date, eight tubes of the 21 tube lot have been eddy current tested and found to be satisfactory. Final processing of the acceptable tubes, consisting of straightening and sizing by pickling, is under way.

The fabrication of 0.005 in. thick depleted uranium foil for breeding gain experiments has been completed. The 0.005 in. thick enriched uranium foil is in the process of being rolled from 0.010 in. thick stock.

All fabrication and jacketing of plutonium-bearing specimens has been completed. The assembly of the special fuel rods, thimbles and baskets awaits delivery of inspected tubing.

C. EBR-II

1. Reactor Plant

During the final checkout of the fuel handling system, difficulty was encountered with rotation of the large rotating seal plug. Sticking was first observed occasionally, but became progressively worse and finally precluded further operation. Preliminary investigation indicated that some sort of mechanical interference had developed between the blade and the trough of the plug "freeze" seal used to seal the primary tank argon blanket gas from the atmosphere.

The seal consists of a stationary, annular trough filled with a low melting point metal alloy within which there rotates a cylindrical sealing blade attached to the plug. For reactor operation, the metal alloy (Cerrotru: 58% bismuth- 42% tin) is maintained frozen at the top and molten at the bottom. For plug rotation, the alloy is maintained completely molten. To aid in establishing the half-frozen condition, the blade is comprised of an upper ring and a lower ring, each with its own set of heaters, and with an insulating gas void between the two. The upper ring is of stainless steel and the lower of copper.

Because the design of the freeze seal for the small rotating plug is similar to that for the large plug, it was decided to remove both plugs for seal inspection. In preparation, it was necessary to remove many of the installations on the plugs, including the cover lifting mechanism, hold-down mechanism, gripper drive, and a substantial amount of wiring. The Cerrotru alloy was also drained from both freeze seals. It was found possible to leave intact all control rod drive installations on the small plug, however.

The large plug was removed at the end of the reporting period, in time to permit preliminary examination of the seal. The apparent cause of the large plug trouble was severe corrosion of the copper (lower) ring by the Cerrotru, particularly in the vicinity of the heaters where the higher temperatures existed. Attack at the heaters was sufficiently severe to completely penetrate the thin copper walls at local points. This probably permitted segments of the copper ring to fall to the trough bottom, catch on projections of the trough, and prevent rotation of the plug. Damage appeared limited to the lower ring of the seal blade and some insulator cans positioned within the trough. The upper ring of the blade and the trough itself were undamaged. No damage was sustained by the small plug seal except for excessive corrosion of the copper ring.

Redesign of the seal for both plugs has been started. A stainless steel lower ring will be employed in lieu of the copper ring. The necessary material has been obtained and rework of both seal blades has been started.

2. Sodium Boiler Plant

An engineering analysis revealed that initial clean-up, cold trapping, and operation of the primary and secondary sodium systems can be performed in a shorter overall time if they are handled individually. To accomplish this, the secondary sodium supply and return lines were cut near the reactor building and a 180° return bend was installed. At the same time, a combination temporary filter and cold trap was installed in the system at this same location. This installation will separate the secondary system from the primary system heat exchanger in the reactor building. This alteration will allow individual operation of the primary and secondary systems during initial cold trapping and systems testing.

Insulation, painting, and installing induction heating wire on pipes and components of the secondary and steam systems continued both in the building and on yard piping.

Installation of iron grating, kick plates, and redesigned mechanical seals in the Dowtherm circulating pumps has been completed. Fabrication is in progress on the support for the new 250 kw heater bundle being installed in the secondary sodium system surge tank. ANL personnel calibrated the level probes for the surge and storage tanks at operating temperatures verifying that they are operational.

Preparations for chemically cleaning the steam system outside the power plant were formalized and a chemical-cleaning contractor was selected. Final preparations for this cleaning were in progress as the period ended.

3. Power Plant

Miscellaneous work, directed toward safer and easier plant operation, continued. This included such items as installing reach rods on valves, and inspection and cleaning of control valves. Work started on painting and color coding of pipe lines associated with the turbine.

4. Fuel Cycle Facility

a. Construction, Installation, and Correction of Construction Deficiencies

(1) Argon System - Both Keystone valves on the argon duct and the Kieley-Mueller freon control valves were reworked by the vendors and somewhat better operation obtained, although there were still some leaks and cooling data indicated erratic opening of the Kieley-Mueller valves. Some of these valves still leak at the body flanges and are being retightened. The small air-argon locks were made leaktight and the mechanical work was nearly completed.

(2) Windows - All 31 shielding window units have been installed. Pairs of removable window slabs are now being installed remotely at the process area side of viewing windows. All of the Argon Cell luminaires and the service sleeve feed-throughs have been installed remotely. Window shutters are being assembled and installed.

(3) Cranes and Manipulators - In-cell cranes and manipulators have been extensively reworked and, except for the bridge drive on the cranes and other minor problems, operation is satisfactory. A complete motion picture record was made, showing remote installation of a manipulator carriage.

(4) Processing Equipment - One of the two melt refining furnaces and the ingot removal and handling device have been installed in the Argon Cell. The furnace was installed using cranes and manipulators in order to verify the remote handling features of this equipment. A dumper which will pour skull oxide into the skull reclamation furnace was received and tested. Operation became erratic when dust accumulated in some of the moving parts. The dumper is being modified. The shielded coffin for transferring fuel assemblies between the EBR-II reactor and the Air Cell is being fabricated by a commercial fabricator and is about 40 percent complete.

The process equipment for refabrication of EBR-II fuel consists of:

1. Radiation resistant machinery for the direct processing of fuel and fuel rods. This equipment is located in the Argon Cell and Air Cell.

2. Auxiliary equipment such as vacuum pressure systems, pneumatic controls and any other components connected directly to the process cell but shielded from it. These items are located in the shielded subcell.

3. Electrical power switching and signalling equipment located in the basement beneath the operator's annulus.

4. The control and indicator stations which will be located in the control annulus.

The operation of the process equipment is from pushbutton stations in the operator annulus, controlling relays, contactors and sequencing devices located in the basement. These, in turn, operate valves in the subcell which supply compressed air or compressed argon gas to the cylinders operating movements on the process equipment.

The installation of refabrication equipment, electrical controls, pneumatic controls, vacuum systems, located in the subcell and basement, has reached the point where these systems can now be checked out. A second man from the Plutonium Fabrication Group was moved to Idaho to assist in checking out the controls for the Argon Cell equipment. Installation of the electropneumatic systems for the Air Cell equipment was started during the reporting period.

(5) Analytical Equipment - A recently obtained gas chromatograph apparatus has been set up in the FCF laboratory and placed into operation for analysis of argon reactor blanket and cell atmosphere gas for minor impurities. Performance on some test gas mixtures appears to be satisfactory.

b. Development of Remote Control Methods and Equipment for Fuel Fabrication - Work has continued on the preparation of installation packets (drawings, installation and operational instructions, material and equipment) for the EBR-II Fuel Cycle Facility. Installation packets were prepared and submitted to Idaho during the reporting period for the Air Cell valve cabinets, control hanger rails, injection casting furnaces and electrical controls, wiring and distribution. Bids were received and an order awarded for the manufacture of in-cell equipment for pulsed eddy current bond testing of the EBR-II fuel rods. The electronic and display equipment for this device is being constructed.

Design drawings were completed, specifications prepared and requisitions submitted for the following items of in-cell equipment:

1. A furnace charging table
2. A furnace mold pallet load and unload station
3. A fuel rod chopper station
4. A sample collecting device
5. A fuel rod assembly station
6. Manipulator tongs for billets
7. Crucible manipulator tongs
8. Injection furnace mold pallets
9. A device for breaking crucible heels

Furnace charge preparation and fuel rod assembly will be master slave manipulator operations. The fuel rod assembly station consists of a device to open the transfer containers used to bring sodium filled jackets into the cell, a holder to present the jackets to the manipulator, a feeder magazine for fuel pins, a turntable for the fuel element magazine and an alignment jig to allow threading the fuel pins into the fuel rods.

5. Instrumentation Development

a. Fuel Element Failure Detector for EBR-II - The data obtained in the experiment simulating the geometry and radiation intensities anticipated for the detector system planned for EBR-II (ANL-6523 Progress Report for May, 1962) has been processed. Commercial instrumentation consisting of three fission counters and one BF_3 counter was used to detect the photoneutrons in the presence of high gamma background flux. The photoneutron energy levels were comparable to delayed neutrons expected to be encountered under actual conditions in EBR-II. By irradiating indium foils, the absolute thermal neutron flux distribution was obtained and the detector efficiency of each counter was calculated.

For making an estimate of the over-all detection efficiency with each detector, an approximate value of photoneutron source strength was used. This was obtained by comparing the flux distribution measurements with the theoretical distribution due to a cylindrical shell source of thermal neutrons in an infinite graphite medium. The absolute strength of the gamma source was determined by a coincidence counting method.

The measured efficiency of the detectors, in conjunction with the integral and differential bias curves obtained, suggests that the best choice of counter would be the one with the highest detection efficiency. This routine conclusion is the direct result of the somewhat surprising discovery that the gamma background does not seem to interfere at all with the operability of any of the detectors tested, even with the rather strong gamma source used in these tests. The neutron source used was at least 100 times stronger than the peak of the burst one would predict from a "catastrophic" failure of one fuel pin. The strong source was required to plot the flux with reasonable statistical accuracy.

A new series of tests will be performed using fission chambers having twice the efficiency, better resolution, but poorer plateau characteristics than the commercial units. A more realistic neutron source strength will be used in the test by reducing the diameter of the heavy water envelope, but the intensity of the gamma source will remain the same. Under these conditions, more realistic neutron/gamma ratios will be available and the tests should indicate whether the more efficient detectors, which have plateaus of much poorer quality, or the less efficient detectors provide the best overall system.

6. Training

Training activities during this period involved reactor technology courses for reactor and fuel cycle technicians, reactor plant systems study, operational exercises utilizing the EBR-I reactor, and a seminar on the EBR-II reactor experimental program.

All reactor technician training was temporarily suspended, for one week, for reactor plant work. Training was resumed when work assignment requirements were determined.

The reactor technology course for new technicians assigned to reactor operations was essentially completed.

A reactor technology course for fuel cycle technicians, intended to provide a broad understanding of the EBR-II reactor facility and its underlying reactor concepts, was initiated. Nine technicians participated for twenty-two class hours. This effort is approximately 50% complete.

Three technicians of the operating organization spent seven working days at EBR-I for familiarization and training in reactor operation. Their schedule included studies of the basic systems and of the operating characteristics of the reactor, completion of startup and weekly interlock check sheets, and supervised startup of the reactor. This training was supplemented with an approach-to-critical experiment using the outer blanket cup, actual calculations, and control rod and cup calibrations. A lecture was also given on reactor stability and the EBR-I meltdown.

A seminar was conducted by the project physicist for the shift supervisors and foremen concerning the EBR-II experimental program.

7. Fuel Development

a. Fast Reactor Fuel Jacket Development - Nb-1 w/o Zr alloy wire is in the process of being fabricated for high temperature fuel jacket development studies. The as-received 0.675 in. diameter bar stock was cold rolled 91% to 0.176 in. square wire, cut in $1\frac{3}{4}$ in. lengths, and annealed at 1100°C for one hour in vacuo. The annealed material was cold rolled another 90% to 0.056 in. square wire and is being cleaned prior to the second anneal at the same time and temperature. The 0.050 in. square wire will be delivered in a 20% cold worked condition.

Two 10 foot lengths of tubing measuring 0.385 in. O.D. x 0.021 in. wall, were received from the Wolverine Tube Co. While inspecting the Nb-5 w/o Zr and Nb-5 w/o Zr-10 w/o Ti alloy tubes, circumferential finish marks were observed rather than the longitudinal marks expected from a drawing operation. Closer examination at 45 and 90X revealed that the tubes were probably belt sanded in an effort to remove the defective outer skin layer of the tubes. Initial metallographic examination of the tubes indicated that the outer layer was severely attacked during pickling or that the outer layer was heavily worked and failed by cracking and tearing.

Even though the tubing is unfit for further fabrication, the material can be used to indicate the behavior of some of the parameters of drawing, such as; the effectiveness of a soft core, resulting diameter to

thickness ratios, and fabricability to 0.156 in. I.D. x 0.009 in. wall tubing. For the fabrication of quality tubing, it will be necessary to remove the defective layer, which might be as much as 25% of the wall thickness, by belt sanding or grinding.

A 10 in. segment of the as-received Nb-5 w/o Zr-10 w/o Ti tubing was cold drawn over a copper core to 0.196 in. O.D., through 13 aluminum-bronze dies. The tubing did not fail and was relatively free from striations. The D/t ratio remained relatively constant. Further reductions to 0.156 in. I.D. x 0.009 in. wall will be attempted through lubricated tungsten carbide dies.

b. Properties of Uranium-Plutonium-Fissium Alloys - The study of the properties of uranium-plutonium-fissium alloys has continued and thermal conductivity data have been obtained on four alloys of high zirconium-fissium content (total Zr, 2.8 w/o). The alloy from the pyroprocessing cycle can be varied in plutonium and fissium content. The different compositions studied reflect the variations in composition that may be expected from the pyroprocessing.

The thermal conductivity values of the four uranium-plutonium-fissium alloys are listed in Table III. The thermal conductivity of the reference alloy, uranium-20 w/o plutonium-10 w/o fissium, is decreased about 8% if the fissium content is raised to 15 w/o fissium or is decreased 20% if the plutonium content is raised to 40 w/o plutonium. However, the values for uranium-10 w/o plutonium-10 w/o fissium and uranium-20 w/o plutonium-10 w/o fissium are practically identical. The thermal conductivities of two alloys remain to be determined - uranium-20 w/o plutonium-5 w/o fissium and the low zirconium uranium-20 w/o plutonium-10 w/o fissium.

Table III. Thermal Conductivity of Uranium-Plutonium-Fissium (High Zirconium) Alloys

(in cal/sec-cm-°C)

Temp. (°C)	U-10 w/o Pu-10 w/o Fs	U-20 w/o Pu-10 w/o Fs	U-20 w/o Pu-15 w/o Fs	U-40 w/o Pu-10 w/o Fs
100	0.043*	0.044*	0.040*	0.029*
200	0.046	0.047	0.043	0.034
300	0.051	0.051	0.048	0.040
400	0.057	0.056	0.053	0.045
500	0.063	0.062	0.060	0.051
600	0.070	0.069	0.067	0.057*
700	0.077	0.077	0.074	0.062*
800	0.084*	0.086*	0.080*	

*Extrapolated values.

The fissium compositions listed in Table IV are calculated for a 20 w/o Pu^{239}-20 w/o U^{235} fuel. Half the technetium is added to the molybdenum and half to the ruthenium. It is also assumed that no zirconium is removed by the processing cycle.

Table IV. Fissium Composition for a Fuel Containing 20 w/o Pu^{239} and 20 w/o U^{235}

	5 w/o Fs	10 w/o Fs	15 w/o Fs
Zr	1.40	2.80	4.20
Mo	1.87	2.75	4.62
Ru	1.98	2.95	4.93
Rh	0.25	0.50	0.75
Pd	0.50	1.00	1.50
Total Fs, %	5.00	10.00	15.00

c. Compatibility of Uranium-5 w/o Fissium with 304 Stainless Steel - The Mark I loading of EBR-II consists of uranium-5 w/o fissium fuel in a cladding of Type 304 stainless steel. A series of heat treatments were performed on diffusion couples of these materials at 550, 600 and 650°C for one, two and four weeks. By microscopic examination of these couples it was determined that bonding did not occur because of oxidation on the bonding surfaces. A new bonding and encapsulation technique has been performed on a new set of diffusion couples. These couples are presently being heat treated at 550, 600, 650 and 700°C for one, two and four weeks. It is hoped that by using this more meticulous bonding method that better results will be obtained.

d. Penetration Rate of Cladding Materials by Molten Fuels - Safety considerations of EBR-II operation require information about the rate of penetration of various prospective clad materials, especially refractory alloys, by molten uranium- andplutonium-containing fuels. The apparatus for testing these materials up to 1500°C in a glovebox is now completed. Using this apparatus tests were run on 304 stainless steel and Armco iron with molten uranium. The 304 stainless steel with molten uranium had been previously tested in another apparatus (ANL-6565 Progress Report for April, 1962). These are our first tests of Armco iron in molten uranium. The preliminary results are indicated in Table V.

Table V. Time (sec) to Penetrate 0.030 in. Wall Capsules of 304 SS and Armco Iron by Molten Uranium

Temp. (°C)	0.030 in. Armco	0.030 in. 304 SS
1138	1.3	0.9
1148	1.1	1.3
1169	1.05	4.3
1187	1.3	7.3
1229	7.0	5.7
1244	5.2	
1300	3.7	4.6

These data are generally in good agreement and serve to confirm the early test results.

8. Process Development

a. Melt Refining Process Technology - A pin cast from melt refined metal from the fifth high-activity-level experiment was exposed to air for 18 days and then exposed to undiluted nitrogen at 650°C for one hour. Its nitridation rate was very low, which is attributed to the formation of a protective oxide coating during exposure to air.

A process is being studied for melt refining uranium fuel pins in an inert crucible under a low melting oxide flux containing uranium dioxide. The possible advantages of this process are higher yield, lower contacting temperature and shorter contacting time. The uranium dioxide oxidizes fission elements whose oxides are less stable than uranium dioxide. An initial experiment had as its objectives (1) finding a flux that is more stable than uranium and has a low melting point and (2) finding a crucible material of sufficient inertness to withstand attack by both the melt and the flux. Preliminary experiments indicate that slip cast beryllia crucibles and a flux base of either 66.5 mole percent BeO-33.5 mole percent BaO which melts at 1430°C or 63 mole percent BeO-37 mole percent SrO which melts at 1380°C are satisfactory crucible-flux base combinations.

Distillation of magnesium-zinc in the melt refining furnace is being studied as an alternative to retorting zinc- and magnesium-coated uranium after the intermetallic decomposition step of the skull reclamation process. In a run in which 400 grams of 50 percent magnesium-zinc was distilled from the crucible, 89 percent of the charge was collected in the graphite condenser resting on the furnace crucible, 8.7 percent was collected on the Fiberfrax insulator inverted over the condenser, and 2.3 percent was not accounted for. The graphite condenser had to be forcibly separated from the crucible after the run. Since such a procedure would not be satisfactory for remote operations, zinc-magnesium distillation will not be feasible unless a satisfactory gasket material is found for use between the condenser and crucible.

b. Skull Reclamation Process - Results of two noble metal extraction runs, performed under inert atmosphere with some exposure of flux to air, gave satisfactory extraction of noble elements, as compared with three runs performed in a dry atmosphere in which noble metal extractions were not effective. The most plausible explanation is that water, absorbed by the deliquescent flux during loading of equipment in air atmosphere, promotes noble metal leaching. The mechanism involved in this action is not yet understood.

Beryllia crucibles made by thixotropic slurry casting continue to perform exceptionally well as containers for the intermetallic precipitation, uranium precipitation, and retorting operations. A 4-inch-diameter 9-inch-high stainless steel-jacketed crucible of this type has successfully undergone the equivalent of ten intermetallic and decomposition steps on a 100-gram-uranium scale. These are the best results obtained thus far with any crucible.

The design of the integrated process equipment for large-scale demonstration ($2\frac{1}{2}$ kg of uranium) of the skull reclamation process is about 80 percent complete. Molybdenum-30 weight percent tungsten alloy has been chosen as the material for transfer lines on the basis of good results obtained in 15 transfers of 8-kilogram quantities of zinc at temperatures up to 850°C. The results of molten zinc transfers under conditions more severe than those expected in skull reclamation indicate that mild steel or stainless steel will be a satisfactory material for waste containers.

c. Blanket Processing Studies - The blanket process involves dissolution of the plutonium-bearing blanket material in a zinc-rich alloy, precipitation of uranium by addition of magnesium, and removal of the supernatant phase whose volume will be reduced by evaporation, if necessary, to a volume suitable for feeding into a future core cycle. The most important factor in determining the efficiency of the uranium-plutonium separation is the efficiency of the phase separation. In thirteen runs at $\frac{1}{20}$ scale (0.5 kg uranium, 5.5 kg magnesium-zinc alloy), the efficiency of transfer was 92.5 to 97.5 percent of the supernatant phase. It is concluded that washing of the uranium precipitate will probably be unnecessary.

d. Plutonium Recovery Process - The distribution coefficients of vanadium, barium, and zirconium between zinc-magnesium solution and molten magnesium chloride at 800°C were determined. Vanadium tended to favor the metal phase at magnesium concentrations of 0.066 to 65.2 weight percent, and barium strongly favored the flux phase throughout the range of magnesium concentrations used (0.77 to 76.2 weight percent). Zirconium showed a preference for the metal phase at all magnesium concentrations tested (0.08 to 75 weight percent magnesium). Some volatilization of zirconium tetrachloride may have occurred during the experiment.

The effect of plutonium concentration on its distribution between magnesium chloride and zinc-10 weight percent magnesium at 800°C was also investigated. As the plutonium concentration was increased from about 0.5 to 5 weight percent, the distribution coefficient increased slightly from about 0.18 to 0.25.

e. Reduction of Thorium Dioxide - Experiments on the direct reduction of thorium dioxide to thorium metal by zinc-five weight percent magnesium alloy have shown that when an argon atmosphere is used

instead of air (see ANL-6573, Progress Report for May 1962, page 25), complete reduction can be achieved over a wider range of flux compositions. The broadened range permits the use of more convenient and economical flux compositions. Other experiments showed that the rate of reduction increases both with increased stirring rate and increasing temperature from 750 to 850°C.

f. Reduction of Uranium Ore Concentrate - The feasibility of reducing uranium ore concentrates by zinc-magnesium-molten flux procedures is being evaluated in a limited program. Uranium metal buttons have been prepared by these procedures. Analytical results on the purity of the product are not yet available.

g. Materials Evaluation - A study is underway to test various types of beryllia for wettability and permeability by molten salts and metals. A sample of isostatically pressed and sintered beryllia which was exposed to zinc at 800°C for 100 hours did not appear to have been wetted, but zinc penetration was noted. Another sample of isostatically pressed and sintered beryllia was exposed to a $LiCl$-$MgCl_2$-MgF_2-$ZnCl_2$ flux at 800°C for 75 hours; some areas were wetted by the flux, which filled nearly all of the pore volume.

A crucible and agitator were tested under skull oxide reduction conditions for a period of 500 hours at 800°C. The charge consisted of 3500 grams zinc, 193 grams magnesium, 154 grams skull oxide (containing approximately 75 percent uranium), and 821 grams of 47.5 weight percent $LiCl$-47.5 weight percent $MgCl_2$-5 weight percent MgF_2 flux. Tested were a tungsten crucible made by isostatic pressing and sintering and a molybdenum-30 percent tungsten agitator which was operated continuously during the run at 500 rpm. At the conclusion of the run, the crucible had undergone no visible attack, but some pitting was noted on the surface of the agitator.

Techniques are needed for fabricating large, impermeable, corrosion-resistant, thermal-shock-resistant crucibles. A study is underway to develop a cement to be used either as a mortar in making brick-type beryllia crucibles or as a component (along with a suitable mineral aggregate) of a mix for making crucibles by the method of concrete casting.

D. FARET

Design studies and measurements are continuing to develop a low conductivity or thermally insulated fuel element for Doppler coefficient studies. An alternate design to the powdered fuel concept described in ANL-6580 (Progress Report for June, 1962) and one believed to be insensitive to thermal cycling consists of ceramic pellets. Each ceramic pellet has a number of small peripheral projections or pads approximately

0.015 in. high which center the pellet within the metal sheath and provide for the required insulating gap between fuel and clad. The tube is gas filled. The laboratory test assemblies of both concepts are shown in Figure 2.

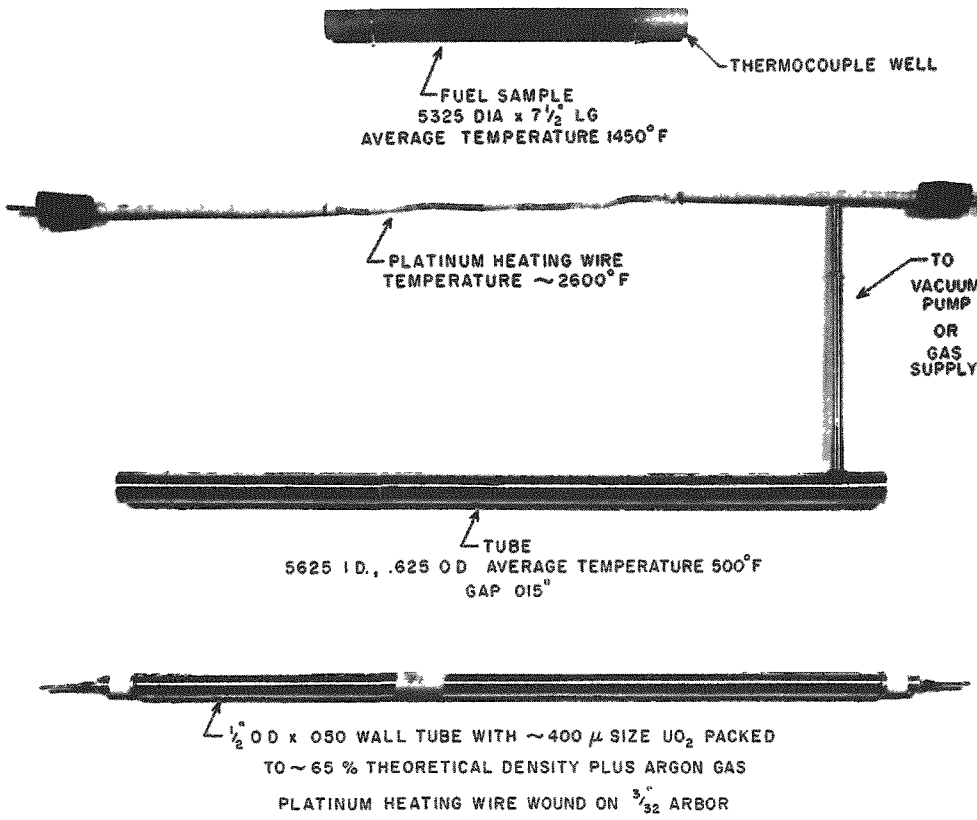

Figure 2. FARET Heated Fuel Samples

The results of tests using electrical heating on the insulated pellets show promise. The results of the latest test are shown in Table VI.

Table VI. FARET Heated Fuel Sample
(0.5325 in. diameter x 7½ in. long)

	°F	°C		°F	°C
Heating wire	2620	1440	Metal sheath	500	260
Center of pellet	1575	860	Gradient across		
Surface of pellet	1445	610	0.015 in. gap	815	435

The heat conduction rate, $\int k(\theta)d\theta$, for this test was 6.9 w/cm which would correspond to a reactor power of approximately 2 Mw for a 1670-liter zoned core. However, it is desired to obtain a gradient across the gap of approximately 540°C which would correspond to approximately 8 Mw of reactor power. It is believed that the required temperature gradient can be demonstrated by experiments now in progress.

The design of the main coolant piping, as shown in the preliminary proposal, dated April 16, 1962, was analyzed for forces, bending moments, and fibre stresses due to thermal expansion and internal pressure.

The results of this analysis showed excessive thermal stresses of 44,000 psi in the reactor-to-heat exchanger line and 37,000 psi in the heat exchanger-to-primary pump line. To stay within the American Standard Code for Pressure Piping (ASA B 31.1-1955) of 31,750 psi at 7,000 thermal cycles from 70° to 1200°F, the reactor exit coolant nozzles has been relocated 90 degrees counterclockwise and the exit pipe redesigned to a U-configuration (maximum stress 31,200 psi). The study is continuing to achieve greater margins between calculated and allowable stresses.

III. GENERAL REACTOR TECHNOLOGY

A. Applied Reactor Physics

1. Neutron Scattering

a. Elastic Neutron Scattering from Iron - The n-γ discrimination detector was used to measure the angular distribution of neutrons elastically scattered from iron for incident neutron energy between 700 kev and 1100 kev. The energy resolution used was 22 kev. Multiple scattering corrections of the angular distributions have not been completed. However, the integral of the angular distribution, which yields the total elastic scattering cross section, is independent of the multiple scattering correction for iron. The total elastic cross section (σ_{el}) together with the total cross section (σ_t) are shown in Figure 3. The absolute cross section was determined by measuring the scattered intensity from iron relative to the scattered intensity from carbon. The multiple scattering correction for carbon has been determined and is included in Figure 3. Comparison of the σ_{el} and σ_t curves in the figure clearly shows the onset of strong inelastic scattering ($\sigma_{inelastic} = \sigma_t - \sigma_{el}$) in the vicinity of 1 Mev.

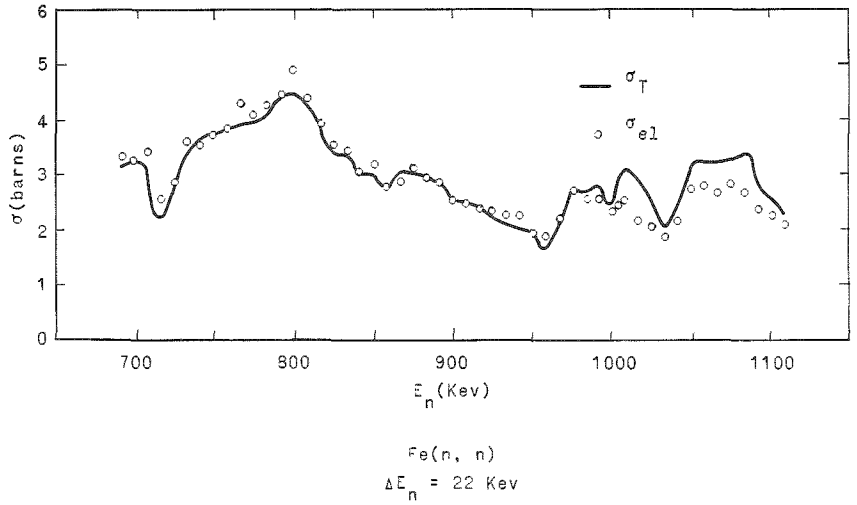

Figure 3. Total and Total Elastic Cross Section of Natural Iron

b. Elastic Scattering of Fast Neutrons by Time-of-Flight - Numerous experimental measurements ($\sim 2 \times 10^3$) were made during this period. The work involved studies of elastic scattering of 700-1500-kev neutrons from antimony, chromium, cobalt, tin, zirconium, copper, niobium, zinc, W^{184}, natural uranium, and tantalum. The neutron energy resolution was approximately 50 kev. The differential scattering cross sections were measured at 10 to 11 angles between 20 degrees and 145 degrees. Absolute normalization was made relative to the known scattering cross section of

carbon. The large amount of data is now being processed in order to reduce the experimental measurements to a compact analytical form. These measurements are a part of the comprehensive study of fast neutron scattering from intermediate and heavy materials.

It is becoming increasingly evident that the above type of work will demand more sophisticated data interpretation and reduction if results are to be obtained with expediency. Efforts to improve the current processing procedure are now in progress with emphasis on the problem of pattern recognition. The experiments themselves involve some hundreds of hours of operation of the accelerator and associated equipment.

2. Cross Section Measurements

The $Co^{58m,g}$, where m refers to metastable state and g refers to ground state, decay curves from the $Ni^{58}(n,p)\,Co^{58}$ cross section measurement reported in ANL-6580 (Monthly Progress Report for June, 1962) have been analyzed to determine the isomer ratio. The results are presented in Figure 4. The rapid rise is the relative yield of Co^{58m} below 1.2 Mev neutron energy is being investigated further.

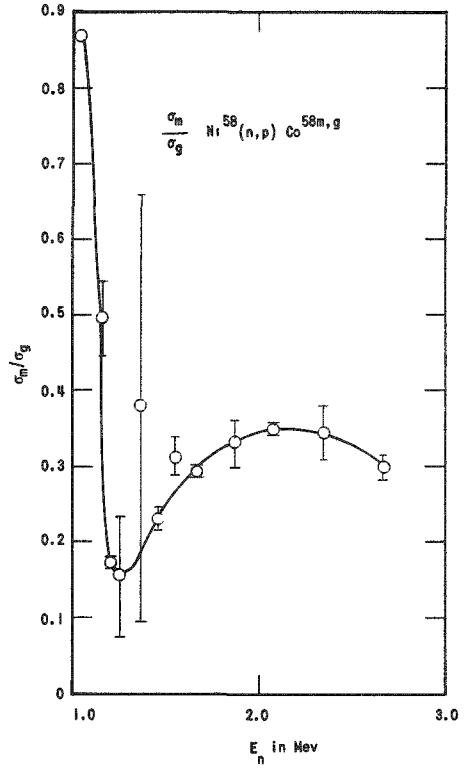

Figure 4. Ratio of Metastable to Ground State Yields for the Reaction Ni^{58} (n,p) Co^{58}.

3. Thermal Critical Experiments (Hi-C and BORAX-V Superheat)

Two markedly different cores were assembled during this period. The first, a continuation of the study of high metal-to-water ratio systems, was made by substituting stainless steel-clad Hi-C, 3 wt-% enriched UO_2 fuel in the central region of the core described in ANL-6580 (Progress Report for June, 1962). Criticality was obtained with 1,875 steel-clad elements in a 1.127-cm triangular lattice forming a 90.6-cm diameter central fuel zone. The surrounding peripheral zone was 11.3 cm thick and included 959 aluminum-clad elements of the same fuel loaded with alternating rows vacant in the same grid. The volume ratios of H_2O/UO_2 were 0.316 and 1.92 in the central and the peripheral zones, respectively. The corresponding atomic ratios (H/U^{238}) are approximately 1/1 and 5.8/1.

A few foil exposures were made in this stainless steel Hi-C core. Completely bare and cadmium-covered traverses were made with dyprosium and gold, and in addition, data were obtained for determination of the cadmium ratio at the center of the core with manganese and copper. Further experiments with this loading were postponed in order to perform some BORAX-V superheat critical experiments.

In the second assembly, the Hi-C core structure was replaced by one which allowed the introduction of a central zone composed of the BORAX-V superheater elements. This was done to obtain flux and power distributions and flooding reactivity measurements for the BORAX-V central superheater. Twelve BORAX-V subassemblies containing 5.18 kg U^{235} and about 180 kg of Type 304 stainless steel were loaded in a 2-4-4-2 array inside four aluminum quadrant cans, forming a central fuel zone approximately 42 cm wide and 61 cm high. This was surrounded by a peripheral fuel zone of the aluminum clad, 3 wt-% enriched UO_2 fuel elements. Approximately 1200 elements were required for criticality when arranged in a square lattice pattern of 1.27 cm pitch. The thickness of this peripheral fuel zone varied from 10 to 25 cm, having an outside boundary radius of about 33 cm. This zone was 122 cm high but moderator height was limited to no more than 91 cm. The volume ratio (H_2O/UO_2) was 1.35, corresponding to an atomic ratio (H/U^{238}) of 4/1.

4. Preparations for Plutonium Critical Experiment

Extension of the Hi-C critical assembly program to include cores containing plutonium as reactivity spikes or for synthetic enrichment has been proposed. The search for a convenient method of varying the plutonium content of the fuel elements has led to a consideration of blending plutonium with UO_2 powder and compacting the mixture in fuel tubes by vibration.

In addition consideration is being given to the use of plutonium-containing glass developed by the Mounds Laboratory. It was anticipated that the plutonium component fixed in the globules of glass would minimize the health hazard in case of fracture of the fuel containers. However, some furnace tests, simulating a power excursion, indicate that substantial amounts of plutonium are released from the glass.

Tests more representative of actual reactor conditions, in that the heat will be developed within plutonium-bearing glass mixed with UO_2 powder, are proposed for performance in the TREAT reactor.

5. Theoretical Physics

a. Mathematical Numerical Methods Analysis - The iterative method for repeated integration of functions of a single variable was mentioned in ANL-6509 (Progress Report for January, 1962) and will be published in the next issue of the Computer Journal. It has been found

possible to combine this approach with the iterative interpolation scheme for functions of several independent variables* to obtain an iterative technique for obtaining repeated integrals and partial derivatives of multivariate functions. The derivation of the algorithm has been completed, and the bivariate case of algorithm is being programmed for numerical tests. Even this, the simplest case, is extremely complicated, and the power of the Algol-60 language and of the Algol-30 compiler for the LGP-30 are of great assistance in this task.

b. EBWR Core Designs with High Neutron Economy - In connection with the study of an EBWR core 2 design consisting of 36 depleated uranium (0.4% U^{235}) elements, D; and 111 enriched (2.7% enriched) elements, E; the feasibility of a 9-rod cold shutdown had been reported based on a distribution of D elements that would break up "local criticalities" (ANL-6433 Progress Report for September, 1961). A uniform distribution of the D and E elements had resulted in a $k_{ex} \simeq 0.07$.

The above results implied a relatively high thermal neutron flux in the D zones, increasing the importance of these zones. It was deduced that buildup of fissionable Pu in the D zones should also have greater reactivity worth than burnup of U^{235} in E zones.

The power developed in these D elements should also be higher than might be expected, due to the high thermal fluxes in the D zones. Furthermore, the power developed in the D zones should also increase with burnup.

The above design considerations were verified by 3 group PDQ-2 calculation on a core using 0.4% enriched D elements and 2.3% enriched E elements. There was a positive reactivity swing of 2.2% which was reduced but not completely at the end of 6800 Mwd/T burnup of E elements.

c. Feasibility of Pu^{239}-U^{235} Cores to Predict Core Dimensions

(1) Fast Critical Experiments - The feasibility of determining criticality using minimum fissile inventories through the assembly of zoned fast reactor systems is being investigated analytically. An analysis was made for a representative case. The critical size of a uranium-fueled assembly is determined by a full size mockup. Then half of the assembly is removed and replaced by a plutonium-fueled region of a sufficient size to again achieve criticality. The difference between the size of the plutonium-fueled part and half the size of a totally plutonium-fueled critical is calculated using multigroup diffusion theory for bare cores. Usually the difference is less than 1%; however, for a case in which the two halves were deliberately

* H. C. Thacher, Jr. and W. E. Milne, "Interpolation in Several Independent Variables," J. Soc. Indust. Appl. Math., Vol. 8, pp. 33-42 (1960).

poorly matched with respect to neutron spectra and diffusion characteristics, the difference was close to 10%. Calculations were also made of the magnitude of errors introduced by uncertainties in the cross sections and in the method of calculation of the total critical size. For all cases considered the error in the former method is only a fraction of that introduced by the latter. Two energy groups were used in all numerical calculations. Machine calculations are now being performed in order to determine how well the results found for bare cores are applicable to radially reflected cores. The comparison of results thus far indicate that the agreement is quite good.

B. Reactor Fuels Development

1. Corrosion Studies

a. Zirconium Alloys for Superheated Steam - The steam testing was begun on a series of experimental Zr alloys which are variations on the Ni-Fe and Cu-Fe ternaries currently of most interest.

These alloys were made by arc melting at Argonne. Operating difficulties caused uncertainty as to purity of the melting atmosphere; thus this series may be remade.

Samples were prepared in duplicate from hot rolled sheets. One sample of each pair was heat treated in an evacuated quartz capsule at 900°C for $\frac{1}{2}$ hour and water quenched. Surfaces were cleaned mechanically and by etching. The final surfaces prior to corrosion test were as etched in HNO_3-HF acid solution, the heat treated samples being etched somewhat more strongly than the others.

The heat treated samples consistently gained less weight than the hot rolled samples of their respective alloys, but this may be a result of the relatively round corners and edges of the former, produced by stronger etching before test, rather than the heat treatment. Usually the heat treated samples were superior to the others at corners and edges after the exposure to steam.

Samples of other Zr alloy stocks of interest were placed in test with the above. Three of the alloys were compacts made from the separate constituent powders by GE and were tested as received. Surfaces were prepared by wet grinding. Two other alloys were made by Carborundum Corporation by arc melting. Hot rolled sheets were made, and samples were prepared similarly to the ANL arc melted specimens.

The surface appearance of all these samples was relatively good, with no definite indication of failure thus far.

These tests are being continued with 22 of the above samples, those which failed have been removed.

b. Lightweight Alloy for Use with Mercury - As reported previously (ANL-6565, Progress Report for April, 1962) good mercury corrosion resistance of four nitrided titanium alloys at 454°C prompted an evaluation of these alloys in mercury at 538°C. In this series of tests, nitrided as well as as-polished Ti-2.5 w/o Al-16 w/o V, Ti-3 w/o Al-5 w/o Cr, Ti-7 w/o Al-12 w/o Zr, Ti-8 w/o Mn and commercially pure titanium were included. However, only the alloys were exposed to both the liquid and vapor phases of mercury. The results of these tests are shown in Table VII.

Table VII. Weight Change (mg/cm^2) of Titanium and Titanium Alloys after 14 Days Exposure in Mercury at 538°C (Static)

	Tested in Liquid Mercury		Tested in Mercury Vapor
Materials	As-polished	Nitrided	Nitrided
(1) Commercially Pure Titanium	-49.14	+0.78	-
(2) Ti-2.5 w/o Al-16 w/o V	-21.47	+0.11	+1.12
(3) Ti-3 w/o Al-5 w/o Cr	-53.90	+0.08	+0.64
(4) Ti-7 w/o Al-12 w/o Zr	-77.56	+0.07	+2.82
(5) Ti-8 w/o Mn	-61.35	+0.13	+0.40

The nitrided alloys show no indication of any significant attack in liquid mercury except a partial disappearance of its golden surface and minor cracks around holes drilled to hang the samples. Round edges showed a remarkable improvement in cracking resistance over sharp edges.

Contrary to the earlier results at lower temperatures (371° and 454°C), the untreated alloys, except for Ti-2.5 w/o Al-16 w/o V exhibited higher weight losses than commercially pure titanium. These results indicate the probable importance of trace elements on corrosion behavior. Future investigations of this problem will be made by preparing alloys at the laboratory. No apparent attack on alloys in the vapor phase of mercury was observed under a low magnification microscope.

Metallographic examination and chemical analyses are in progress. Evaluation of these materials under dynamic conditions is planned.

2. Ceramic Fuels

a. Uc-PuC Fuel Development - Work continued on the uranium monocarbide precipitated from liquid metal media and supplied by Chemical Engineering Division. Earlier work on sintering of this material indicated

that when specimens were held at 1300°C for one hour before raising the temperature to a sintering temperature of 1750°C the time at temperature had little effect upon increasing the bulk density. This would indicate that densification was occurring below 1750°C, possibly at 1300°C or lower. As a result of these sinterings, it was thought desirable to determine the shrinkage versus the temperature in a vacuum for some of these carbides. This was done in a vacuum dilatometer maintaining as nearly as possible, a rate of temperature rise of 38°C/10 minutes from 750°C to 1500°-1600°C, the termination temperature. The specimens used were $\frac{3}{8}$ in. in diameter by $\frac{1}{2}$ in. long and were formed isostatically. Four different compositions were run: three were precipitated material and the fourth was material supplied by the United Nuclear Corporation, made by the reaction of propane with hydrided uranium metal.

The specimens made from precipitated carbides produced shrinkage versus temperature curves which were very similar. Shrinkage began very slowly at 750°C and increased until at 1200°-1300°C it was very rapid. At 1300°-1350°C the shrinkage began decreasing and at 1450°-1500°C it virtually stopped. The specimen made from the propane-metal reaction began to shrink linearly at 950°C; at approximately 1150°C the shrinkage abruptly increased and continued at an almost linear rate to 1600°C which was the termination temperature.

In each of the shrinkage runs for specimens of precipitated carbide there was a marked evolution of gas beginning at approximately 950°-1000°C. The pressure increased from less than 5×10^{-5} mm to greater than 5×10^{-4} mm at 1200°-1250°C. Above this temperature the pressure fell off. The specimen made from the gas-metal reaction also evolved gas through this temperature range but the gas evolution was not nearly so large as with the other specimens.

These data indicate that shrinkage and the accompanying reactions involving oxygen start at temperatures much lower than originally had been thought. Sintering schedules will have to be designed for these carbides with a slow rate of temperature rise through this temperature range, even if carbides are produced by the precipitation technique which have considerably lower free carbon and oxygen contents.

The abrupt change in shrinkage rate which occurs at about 1150°C with the specimen of gas-metal reacted material evidently accompanied the melting of uranium metal. Metallographic examination of this specimen, after the shrinkage run, revealed the presence of uranium metal which had migrated to the surface of the specimen.

Chemical analyses are being done on carbide samples heated to various temperatures in an effort to determine exactly what reactions are taking place through this temperature range. When the equipment is available, thermogravimetric determinations will be made for some of these carbides over the same temperature range.

b. Uranium Phosphide - The objective of this study is to characterize the various compounds in the U-P system and evaluate them as possible reactor fuel materials.

Work to date has consisted of preparing experimental quantities of uranium phosphides by direct reaction of the elements. While synthesis by gas reaction is also feasible, reaction of the elements was thought to offer several advantages. Other methods of producing uranium phosphides will be examined in the future.

Various proportions of the two elements were mixed and pelletized. Use of finely divided uranium metal, reduced by hydriding, necessitated performing all the operations in a glovebox containing an inert atmosphere. Samples were fired in a quartz tube having a low partial pressure of helium. A thermocouple was mounted against the bottom of the molybdenum crucible to indicate the beginning of the reaction. The reaction was strongly exothermic and began in the range 300°-400°C, depending on the U/P ratio of the starting mixture. Temperatures estimated to be as high as 1500°C were reached in fractions of a second, falling off as sharply to the furnace ambient temperature. A yellow modification of phosphorus condensed on the tube in all runs. X-ray analysis revealed the following phases to exist (in decreasing order of abundance):

Results of Uranium Phosphide Synthesis Experiments

Run No.	Starting Mixture	Phases Present
1	U:4P	UP_2, U_3P_4, UP and UO_2
2	U:2P	U_3P_4, UP, UP_2 and βUO_2
3	U:P	UP, βUO_2 and U_3P_4

The material from Run No. 3 was predominantly UP, containing less than 5% of other phases. It has metallic luster and preliminary data show it to have a melting point above 2460°C. The presence of UO_2 in all of the samples may have been a result of the grade of phosphorus used. A higher purity red phosphorus has been obtained and will be used in future experiments. Larger quantities of the three compounds will be produced for the purpose of determining some of their properties.

3. Fabrication Development

a. Fabrication of Zirconium Alloy Tubing - The program objective is to develop methods of fabricating high quality tubing in size ranges not commercially available. Work to date has been limited to development of techniques for fabrication of tubing smaller than $\frac{1}{2}$ in. O.D.

Investigation of the copper plating procedure reported in June is continuing. The versatility and dependability of this procedure is being studied to prevent die seizing when Zircaloy-2 tubing is drawn through carbide dies.

b. Fuel Fabrication Facility - Installation of the Extrusion Press - A gloveboxed, 600-ton, direct-indirect horizontal extrusion press, a detwisting stretcher straightener and auxiliary equipment are being installed in Building 350. Because of the large size of the equipment, it was necessary to cut an opening through the east wall to move it into the area. Activities have of necessity been curtailed in the building and as a result necessary maintenance and overhauling of equipment is being accomplished.

4. Irradiation Studies

a. Postirradiation Examination of Blister Area in a CP-5 Fuel Element - Late in 1961 a defect developed in one of the CP-5 fuel elements. The elements are made of concentric tubes of Al-17.5 w/o U-2 w/o Ni alloy clad with X8001 aluminum alloy. The first indication was abnormally high fission product activity in the helium cover gas system. The defective element was subsequently identified by sampling the gas over each element as it was isolated in a storage hole.

After approximately a five month cooling-off period, the subassembly was examined and a circular ring of white scale with a dark area in the center was found on the O.D. of the inner tube while a blister directly opposite this white area appeared on the I.D. of the same tube. The tube was then subjected to sectioning operations in order to examine these areas more closely.

The size of the blister area as determined with a metallurgical microscope was approximately $\frac{1}{2}$ in. and 1 in. at its smallest and largest dimension respectively. An examination of this same area revealed that water had penetrated through the cladding and reacted with the fuel, resulting in an oxide core. The only remaining portions of fuel were attached to the outer edges of the cladding and no reaction layer was evident on the core surface. The core appeared to have been stripped from the cladding in the remainder of the sample and there was no evidence of a high temperature reaction on the clad, indicating that corrosion rather than melting was the cause of core removal.

The absence of any abnormal attack on the outer surface of the cladding at areas immediately adjacent to the defect and the rounded appearance of the cladding inside the blister area, compared to the relatively level condition of the cladding directly opposite, suggests that the reaction was proceeding outward to ultimate failure. Since no swelling or corrosive effects could be found in the white circular area immediately opposite the

blister, it is probable that this is not a defect but, rather, a manifestation of the high heat flux that would result from presence of the blister on the opposite side.

The absence of any abnormal corrosion eliminates this factor as a significant contributor to the failure. The decreased thickness of the cladding at the blister indicates that a hot spot might have developed from increased concentration of fuel at that point, but it is more likely that this is a direct result of the inner core reaction. There was apparently no effect upon the swelling related to any abnormality in the structure of the fuel, as evidenced by an examination of the core next to the blister and near the top of the tube.

The results of the examination did not reveal by which mechanism the blister occurred because the catastrophic nature of the incident erased all vestiges of contributors to the failure. It could be speculated that the failure was caused by either a point defect on the cladding which allowed water to penetrate into the core with a subsequent water reaction followed by swelling, or by some internal defect such as decreased cladding thickness or increased fuel concentration which resulted in a hot spot, softening of the matrix, blistering, penetration of the cladding at the weakest point and an ensuing water-fuel reaction.

b. Postirradiation Annealing of Thorium-Uranium Alloys - The postirradiation annealing experiments on thorium-uranium alloys described in ANL-6580 (Progress Report for June 1962) have been continued to higher temperatures. The information obtained to date is summarized in Table VIII.

Table VIII. Cumulative Volume Increase in Irradiated Thorium-Uranium Alloy Specimens after Annealing for 100 Hours

Specimen No.	Composition, w/o	Total Atom Burnup, a/o	Annealing Temp., °C	%ΔV* a/o Burnup	Specimen No.	Composition, w/o	Total Atom Burnup, a/o	Annealing Temp., °C	%ΔV* a/o Burnup
ANL-42-7-1	Th-10 U	4.0	550	0.2	ANL-42-1-3	Th-20 U	4.5	550	0.2
			600	1.0				600	0.7
			650	1.9				650	1.5
			700	2.0				700	1.5
			725	2.6				725	2.5
			750	2.7				750	3.2
			775	3.7				775	3.0
ANL-42-1-2	Th-15 U	4.5	550	0.3	ANL-42-5-4	Th-25 U	4.2	550	0.4
			600	0.6				600	1.5
			650	0.7				650	2.1
			700	0.9				700	1.8
			725	1.6				725	2.8
			750	1.7				750	2.7
			775	1.9				775	3.2

* The volume change is based on the assumption of no weight change and was calculated from the expression $\%\Delta V = \left[\left(\frac{\rho_i}{\rho_f}\right) - 1\right] \times 100$ where ρ_i and ρ_f are the initial and final densities respectively.

None of the specimens have begun to swell catastrophically, even after 100 hours at the highest annealing temperature of 775°C. These results confirm the irradiation experiments, which showed no pronounced swelling during irradiation under temperatures that ranged up to 770°C.

The annealing experiments have been temporarily discontinued after the anneal at 775°C. The ends of the specimens are being metallographically polished in order that replicas may be taken for optical and electron microscopy. During this interval the annealing apparatus will be modified in order to monitor the release of krypton-85 from the specimens.

5. Properties of the Thorium-Uranium-Plutonium System

As part of the study of the thorium-uranium-plutonium system a series of density measurements of binary, arc-cast, nonequilibrium thorium-uranium alloys has been made. The measurements were made in a glovebox in preparation for subsequent measurements on binary plutonium alloys. Density measurements on plutonium alloys have been made in the past to an accuracy of about plus and minus three parts in one thousand. On the other hand, density measurements made at the National Bureau of Standards on nonradioactive materials are reported with an accuracy of plus and minus one part in 100,000 for materials like stainless steel with a density of about 7.9 and with an accuracy of one part in 10,000 for a material like platinum with a density of about 21.5. The difference is due to the circumstances and conditions under which all measurements in gloveboxes are made and is typical of the problems that arise when data on plutonium alloys is to be obtained. Density determinations in gloveboxes have been made to an accuracy of about plus and minus three parts in 10,000.

Hydrostatic weighing methods and a buoyancy method, using monobromobenzene as a liquid have been employed. Corrections were applied for buoyancy effects, density changes of the monobromobenzene with time, pressure fluctuations and temperature variations in the glovebox and liquid. Dry nitrogen atmosphere was used in the glovebox instead of air.

The results of the measurements of a series of arc melted thorium-uranium alloys are shown in Table IX. A plot of the specific volume vs composition in weight percent, Figure 5, gives the straight line relationship which is expected from binary two phase alloys. Since the solubility of thorium in uranium is small, the point for the density of pure uranium falls onto the straight line. This is not the case on the thorium side where the phase diagram indicates that up to 10.5 a/o or 10.7 w/o of uranium are soluble in solid thorium at 1265°C. The solubility decreases to less than 0.7% at room temperature. Since we know from X-ray diffraction studies the effect of uranium alloying additions to thorium on its lattice parameter, we can deduce from a large scale plot, Figure 6 that under the nonequilibrium cooling conditions prevalent in our arc melted buttons about 1.4% uranium are retained in solid solution.

Table IX. Density of Arc Melted Thorium-Uranium Alloys

Number	Alloy (Nom %) Th	U	Density g/cm³ at 25°C	Mean Density
B751	100	-	11.6793	
			11.6763	11.678
B752	90	10	12.1130	
			12.1120	12.113
B753	80	20	12.6039	
			12.6082	12.606
B754	70	30	13.1550	
			13.1629	13.159
B755	60	40	13.7620	
			13.7643	13.763
B756	50	50	14.4390	
			14.4367	14.438
B760	0	100	19.0003	
			19.0157	19.013

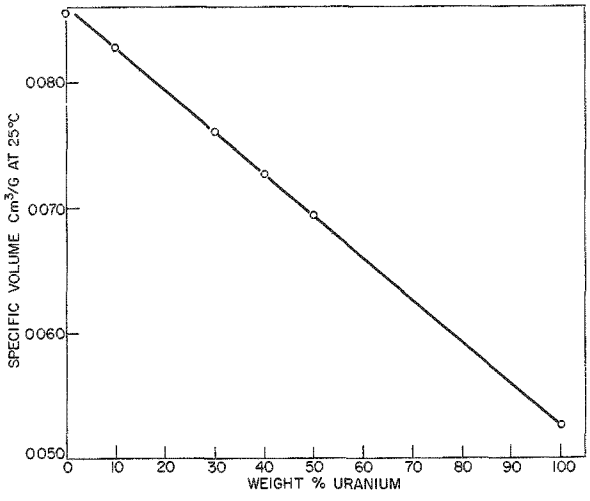

Figure 5

Specific Volume of Thorium-Uranium Alloys. Arc Melted Buttons Made from Commercial Thorium and Dingot Uranium.

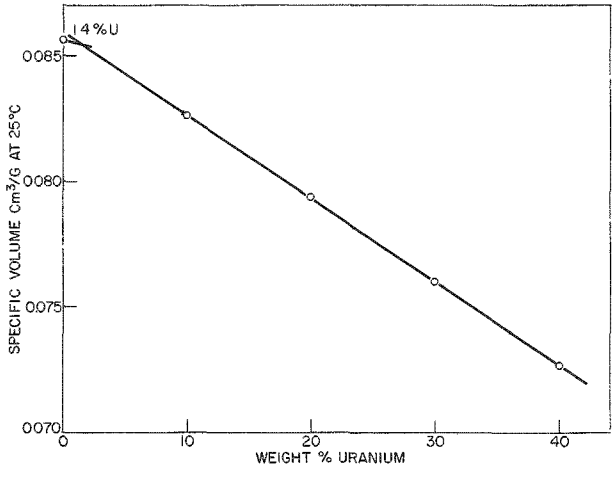

Figure 6

Specific Volume of Thorium-Uranium Alloy (Large Scale Plot)

6. Nondestructive Testing

a. Ultrasonic Techniques

(1) A Study of the β to α Transformation in Pu - Work has continued on the project begun in June to measure velocity and attenuation in plutonium. Various combinations of crystals and various electrical parameters have been tried to obtain a good signal in a highly attenuating specimen using uranium in place of plutonium. The proper experimental setup has been worked out for the uranium specimen, the subsequent stage being the actual measurement of sound velocity and attenuation on a plutonium specimen in a glovebox alloted for this purpose.

A plutonium specimen $\frac{5}{8}$ in. in diameter and $\frac{1}{8}$ in. thick is being prepared for this experiment. Twenty mc ultrasonic energy will be used for the time delay measurements. Further improvements in the method of generation and detection of the sound undoubtedly will be made but the present equipment seems to be adequate for the purposes of the experiment.

(2) Imaging Studies - An ultrasonic imaging technique employing an ultraviolet excited phosphor whose luminescence is quenched by ultrasonic radiation has been briefly studied. This technique appears to be appreciably less sensitive than photographic methods because it was necessary to use lenses to concentrate the ultrasonic energy in order to obtain sufficient intensity for detection. A threshold sensitivity in the order of 5 to 10 watts/cm^2 appeared necessary.

The use of photographic paper exposed to ultrasound in a developer solution has also been studied. The action of the ultrasonic radiation is to accelerate the development of the paper. Although the threshold sensitivity of this technique seems to be in the same order of magnitude as that of other photographic methods, it is much faster than the other film techniques being used. Exposure times in the order of 20 to 30 seconds are common for the photographic paper technique as opposed to 5 to 10 minutes for the photographic film method. Further studies of this imaging technique employing metal test objects are now in progress.

The inspection facility employing the continuously pumped, electronically scanned ultrasonic pickup tube at Northwestern University has been modified so that test objects can be inserted into the ultrasonic beam in an easier manner. Initial observations indicate that 3 to 5% thickness variations in 1 in. aluminum can be detected by visual observation of the kinescope presentation. Further studies with other test objects are now in progress.

Attempts to seal quartz piezoelectric windows on glass tubes for use in a sealed-off ultrasonic pickup tube are now under way. Although no completely satisfactory seals have been made as yet, partial successes with soldering techniques have been encouraging.

b. Lamb Waves - The last series of tests comparing Lamb Waves and shear waves in small diameter thin-walled tubing were completed and analyzed, (ANL-6516 Annual Report for 1961). These investigations were concerned with I.D. transverse and longitudinal simulated cracks in 1100 Al tubing having an O.D. of 0.5 in. and a wall thickness of 0.032 in. Simulated transverse cracks were made on the I.D. wall by means of a special device which fits into the tube. These defects subtend a central angle of 60°.

It was found that with longitudinal flaws, a defect signal was received not only at a Lamb or a shear wave angle of incidence but at any angle of incidence; the amplitude of the signal was constant. The position of the transducer is such that sound does not enter at d, the thickness of tube wall, but rather at some value of thickness larger than d. This difference in d is a function of the angle of incidence. This factor coupled with the beam spread of the unmasked transducer gives a range of fd products and the angles of incidence at one position of the transducer. As a result there is more than one type of wave motion in the tube at a given angle of incidence, and there is propagation regardless of angle of incidence. Power limitations of the reflectoscope used did not permit masking and collimating the beam; this might eliminate this problem entirely.

For I.D. transverse defects, it was found that, in general, defects less than 0.001 in. in depth were located using Lamb waves but could not be located with shear. Defects greater than 0.001 in. were readily and easily located by both methods.

c. Neutron Techniques - The neutron beam facility at JUGGERNAUT reactor is now in operation. The neutron intensity at the reactor face is in the order of 10^7 n/cm^2-sec at a reactor power level of 200 KW. The beam is confined to about a $2\frac{1}{2}$ in. x 4 in. rectangular area and is reasonably uniform in intensity within this area. The gamma intensity in the beam is low enough to permit the use of direct exposure techniques.

The construction of a concrete block shielding wall eliminated much of the high radiation background initially encountered, and permitted the beam catcher to be moved away from the reactor wall. This permits more flexibility in the exposure arrangements. Further improvements now in use include the operation of a fission counter as a beam monitor and the installation of a remote shutter operation control.

The beam is presently being used to evaluate the potential usefulness of several transfer exposure techniques which could not be used with the lower neutron intensity available at the radiographic facility in the CP-5 reactor. Additional data are also being taken to compare the relative photographic speeds of several techniques in the two different neutron beams.

C. Heat Engineering and Fluid Flow

1. Axisymmetric Free Convection Heat Transfer Study

The axisymmetric laminar free convection heat transfer study of a liquid metal flowing along a thin vertical cylinder has been reported in ANL-6473 (Progress Report for November, 1961). The Karman-Pohlhausen integral method for the solution of the boundary layer equations has been used. It was concluded at that time that the integral method was not reliable for small values of the parameter

$$\xi = 2^{3/2}\, x/r_0\, Gr_x^{1/4} \quad ,$$

where x is the axial distance along the pin, r_0 is its radius, and Gr_x is the Grashof number based on x as the characteristic dimension. The analysis has been extended to obtain more reliable heat transfer results for small values of the parameter ξ. To this end, the perturbation technique is being employed to obtain the solution of the boundary layer equations.

2. Fast Reactor Test Facility Heat Transfer

The thermal conductivity of powder fuels calculated by the method of Kunii and Smith[1] indicates that particle sizes on the order of 0.02 cm and smaller are required to minimize the contribution due to thermal radiation. However, it was found that this method yields results which are not in good agreement with much of the data existing on powders in this size range. Although the discrepancies may be accounted for by the fact that most of the data was based on blends of mixed particle sizes, a more consistent correlation is presented by Laubitz.[2] This correlation is substantiated very well by the Laubitz data for various ceramic particles (mean diameters from 0.01 to 0.04 cm) in air to 1100°C, reasonably well by some NACA data on UO_2 in argon, but consistently overpredicts other data on fine silicon carbide powder in air and helium.

On the basis of the Laubitz relation it is estimated that the effects of thermal radiation in UO_2 or UC powders will be nearly eliminated for particle sizes 0.01 cm or smaller, even at 1200°C. Particle size of this order may also reduce the "wall effect" of the fuel tube on conductivity to a point where it may be ignored. It also appears that the effect of the relatively high conductivity of UC is substantially diminished in the powder form, and for 0.01-cm particles in argon the conductivity of both UC and UO_2 is estimated to range linearly from approximately 0.0024 w/(cm)(°C) at 0°C to 0.0075 w/(cm)(°C) at 1200°C. This would allow 60% dense fuel rods of 1.2 cm diameter to reach a central temperature of 1200°C (with 400°C coolant) at a power density of about 90 kw/liter of fuel, a power density of interest to the FARET Driver-Buffer Concept.

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1 Daizo Kunii and J. M. Smith, "Heat Transfer Characteristics of Porous Rocks," AiCH.E. Journal 6, No. 1, 71-78 (1960).

2 M. J. Laubitz, "Thermal Conductivity of Powders," Canadian Journal of Physics, Vol. 37 (1959).

D. Chemical Separations

1. Fluidization and Volatility Separations Processes

a. Fluoride Separations - The separation of mixtures of the hexafluorides of uranium and plutonium by thermal decomposition has been further tested in several experiments. The uranium concentration of the solid plutonium tetrafluoride produced by thermal decomposition of hexafluoride mixtures at 300°C increased with the residence time of the mixture in the vessel. When hexafluoride mixtures containing greater than 90 percent uranium were circulated through a vessel at 300°C, solid plutonium tetrafluoride was produced with a uranium content varying from 0.091 to 1.3 percent for residence times of 0.88 and 6.6 minutes, respectively.

Six preliminary experiments were carried out on the thermal decomposition of uranium hexafluoride vapor at about 330°C. The initial pressure of uranium hexafluoride in each experiment was 50 mm at 300°K. The reaction time in each case was five hours. All of the experiments were carried out in the same nickel vessel in which the products of the reaction were allowed to accumulate. The integral decomposition rates decreased throughout the set of experiments from 0.075 to 0.020 percent of the initial uranium hexafluoride consumed per minute. The ratio of moles of residual gas produced to moles uranium hexafluoride consumed decreased from 0.61 to 0.35 during the series. The decrease in rate may be due to side reactions occurring such as uranium hexafluoride reacting with the nickel vessel.

b. Direct Fluorination of Uranium Dioxide Fuel - A $1\frac{1}{2}$-in. fluid-bed apparatus has been built for laboratory studies of the fluorination step of the Direct Fluorination Process. The apparatus will be used to fluorinate pellets of uranium dioxide containing plutonium dioxide and fission product element oxides.

Fluorinations of crushed uranium dioxide pellets with and without the presence (for heat removal) of an Alundum bed were made. A study was made of the reaction rate for uranium dioxide pellet fragments of sizes $\frac{1}{8}$-in. and larger and for beds containing crushed pellets and fines. As expected, the presence of fines in the crushed pellet charge materially increased the reaction rates. Declad pellets would probably consist of fragments and fines rather than whole pellets. Because of the small void space in a packed bed consisting of these fragments, fluidization of the inert alundum bed in the interstices would be inhibited and this would make good temperature control difficult. To overcome this problem, a scheme is proposed in which the pellets are oxidized prior to the fluorination step. The oxidation of uranium dioxide produces U_3O_8 which is in the form of fines. When the uranium oxide is in the form of fines, good fluidization is possible, hence a more uniform reaction rate should be obtained. Oxidations of crushed pellets were carried out with and without an inert bed. The rate of

oxidation with the presence of an inert bed at 450°C using 20 volume percent oxygen in nitrogen was found to be faster by a factor of three than the fluorination reaction and would therefore be adequate for process use. Elutriation of the U_3O_8 fines from the Alundum bed was found to be independent of the rate of formation of U_3O_8. When the oxidation reaction was carried out without the presence of an inert bed, the U_3O_8 fines formed were not carried out of the reaction zone but rather remained as a compact cake.

A new series of pilot plant-scale runs is underway in continuing studies designed to optimize process conditions in the direct fluorination of uranium dioxide pellet fuel. In these runs, additions of pellets were made during the run to keep the pellet bed height constant in order to establish the effect of other process variables on steady-state fluorination behavior. Steady-state fluorination is defined for this purpose as an operating period of at least four hours during which operating conditions, including uranium hexafluoride production rates and fluorine efficiencies, remain constant. Continued operation by this procedure results in a size distribution of residue pellets, a condition probably comparable to having a feed of cracked and broken pellets.

In three runs using 6-in. pellet beds at 500°C with 10 percent fluorine in nitrogen (no oxygen or recycle), the effect of gas rate on uranium hexafluoride production rates was measured in steady-state periods for three total gas velocities of 0.5, 1.0, and 1.35 ft/sec. In these runs the production rates increased with about the $\frac{1}{3}$ power of the gas velocities.

Two runs were made to determine the effect of varying oxygen contents of the fluorine feed on fluorine efficiency and uranium hexafluoride production (see Table X). Caking tendencies were shown in the 13 percent oxygen run (Run 60) but not in the 5 percent oxygen run (Run 59). Further runs are planned to define satisfactory conditions of pellet bed height and oxygen concentration.

Table X. Results of Uranium Dioxide Pellet Fluorination Runs Using Oxygen in the Fluorine Feed Without Recycle

Alundum Bed Height:[a] 24 inches
Pellet Bed Height: 6 inches
Temperature: 500°C
Fluidization Velocity:[b] 1 ft/sec

Run No.	Duration (hr)	Inlet Gas Composition (%)			Fluorine Efficiency[c] (%)	UF_6 Production Rate [lb/(hr)(sq ft)]
		F_2	O_2	N_2		
59	5	20	5	75	53.5	37.2
60	2	15	13	72	75.6	40.2

[a]Based on empty column.

[b]At operating conditions.

[c]Fluorine efficiency informing uranium hexafluoride.

Supporting studies in mock-up systems are planned to examine some of the effects of uranium oxide fines in fluidized-packed bed systems.

c. Plutonium Pilot Plant Facility - Work on the installation of the plutonium pilot plant facility is proceeding. Final corrections of work on the "phase one" room modification contract are being completed. Preparations for installation of completed items of process equipment (including the fluorinator) are being made. The "phase two" contract for service installations which involves almost all construction work except that done inside the boxes is being prepared by Plant Engineering. Current Critical Path Analysis indicates a final completion date in the middle of 1963.

d. Separation of Uranium from Zirconium Alloy Fuels

(1) Studies of Chlorination and Fluorination Steps with Down-Flow Fixed-Bed Filters - Current work on the two-step chlorination-fluorination scheme for reprocessing uranium-zirconium alloy in the $1\frac{1}{2}$-inch-diameter fluid-bed column is concerned with further study of the effect of the use of phosgene in the chlorination step on overall uranium behavior in the system. Phosgene was fed with hydrogen chloride in a 1:4 ratio during the 5-hr initial reaction period (chlorination) and then used alone for an additional period of 3 hr prior to fluorination. All of the reactions were conducted at 400°C. For a series of runs conducted (atypically) without the static-bed filter in place but using the empty filter section (also maintained at 400°C) as a "settling" chamber, results indicate that uranium losses, as detected by the uranium concentration in the zirconium tetrachloride condenser, were significantly higher with the introduction of phosgene as a chlorinating agent. Typical data showed a loss of 0.83 weight percent of the original uranium charge when hydrogen chloride alone was used whereas losses increased to 14 weight percent when phosgene was used after hydrogen chloride and increased further to 44 weight percent when a mixture of hydrogen chloride and phosgene was used followed by an off-gas phosgene treatment. Results also showed that losses to the off-gas water scrubber (a packed unit down-stream of the zirconium tetrachloride condenser) increased with increased duration of the hydrochlorination period indicating this reaction period should be minimized.

Investigation of the causes for halogen ignitions encountered upon venting the uranium hexafluoride cold traps after the fluorination steps preparatory to hydrolyzing is continuing (see Progress Report, June 1962, ANL-6580, page 44). Preliminary data from infra-red analysis of vapor samples taken from the cold traps at -80°C, 0°C, and at room temperature show the presence of chlorine trifluoride which could be a contributing factor to the ignitions. Revised trap purging and handling techniques are being instituted, also pretreatment with hydrogen fluoride will be used to expel the major portion of chlorine before fluorination and thus minimize the possibility of forming mixed halogens.

(2) Fluid-Bed Hydrolysis of Zirconium Tetrachloride - Fluid-bed hydrolysis studies of the conversion of zirconium tetrachloride to the dioxide by reaction with steam were continued in the 6-inch diameter column (see Progress Report, May 1962, ANL-6573, page 39). The effect of process variables is currently being measured by the quantity of fine material (-200 mesh) produced during a run as opposed to the fraction remaining in the bed particles as an adherent coating. A base-line attrition run on a zirconium oxide-coated aluminum oxide bed showed that about 5 percent of -200 mesh material was produced in a 6-hr period at 350°C with no tetrachloride feed and 2.5 scfm fluidizing gas rate. The quantity of fines found in the final beds in other runs which investigated bed temperature (250 to 350°C), steam excess (2.1 to 5.3 times the stoichiometric requirement) and zirconium tetrachloride feed rate (1.8 to 3.3 kg/hr) was found to be affected only by higher feed rates in that higher rates resulted in greater fines production (the maximum value at 3.3 kg/hr feed rate was 15.9 percent of -200 mesh material for a 3-hr run using fresh Al_2O_3 fluid bed). Satisfactory operation (no pressure buildups) of the unit prevailed with the continued use of high filter blowback air pressure (80 psig as compared to 30 psig used previously).

2. Chemical Metallurgical Process Studies

a. Chemistry of Liquid Metals - Thermal analysis of 7.4 and 60 atom percent tantalum-zinc alloys gave thermal arrests at about 600, 670, and 760°C. The zinc-tantalum system apparently consists of at least five intermetallic phases which decompose peritectically. The results of an X-ray examination of zinc-tantalum compacts having a wide range of zinc to tantalum atom ratios also indicate the existence of at least five zinc-tantalum intermetallic compounds.

An experiment was performed to establish the stoichiometry of the zirconium-iron precipitate which is formed when iron is added to a solution of zirconium in liquid zinc. It was found that the precipitate contains approximately two atoms of iron to one atom of zirconium, although the data do not indicate whether or not the precipitate also contains zinc. It was found that zirconium is quantitatively removed from liquid zinc by the addition of iron.

The solubility of zirconium in liquid magnesium-zinc solutions of nominal 20 and 30 weight percent magnesium content has been measured as a function of temperature. In 20 weight percent magnesium-zinc solution, the solubility varies from 1.6 to 0.02 weight percent zirconium at temperatures of 758 to 584°C, while in a 30 weight percent magnesium-zinc solution it varies from 0.9 to 0.002 weight percent zirconium at temperatures of 764°C to 482°C. These values may be compared with the values of 0.49 weight percent zirconium at 709°C and 0.02 weight percent at 422°C in 50 weight percent magnesium-zinc solution which were reported in Progress Report for February, 1962, ANL-6525, page 48. The existence of several equilibria intermetallic phases is indicated.

Metallographic examination of zinc-magnesium-uranium alloys has revealed the existence of a ternary solid phase. A sample of this new phase has been isolated for chemical analysis.

The magnetic susceptibility of two additional samples of UCd_{11} was measured over the temperature range 2.3°K to 296.2°K (see Progress Report for March 1961, ANL-6343, page 76 for preliminary data). The new data show Curie-Weiss behavior [$\chi = c/(T - \Delta)$] with $C = 1.77_8$ and $\Delta = 41.2$°K from 4.2°K to room temperature. The Bohr magneton number for the uranium is calculated to be 3.78. The magnetic data may be interpreted on the basis of an $5f^2$ ($^3H_4$) configuration for uranium in the compound. A comparison of measured results with theoretical equations indicates that the uranium atoms are perturbed by a crystalline field of cubic symmetry. The overall splitting of the $^3H_4$ multiplet caused by this perturbation is approximately 221 cm^{-1}. The new data on the magnetic succeptibility of UCd_{11} is considered to be more reliable than the previously reported data.

Experiments with a new effusion cell with small orifice area have shown the following intermediate phases in the praseodymium-zinc system: $PrZn_{11}$, Pr_2Zn_{17}, $PrZn_{4.3}$, $PrZn_{3.5}$, $PrZn_2$, and PrZn. The use of the new cell resolved the two solubility ranges $PrZn_{3.4-5.6}$ and $PrZn_{6.9-7.4}$ reported previously (see Progress Report for August 1961, ANL-6409, page 57) into the line compounds $PrZn_{4.3}$ and $PrZn_{3.5}$. These results are in good agreement with X-ray data.

b. Metal Distillation Studies - The induction coil for the large-scale cadmium distillation unit was plasma-arc sprayed with aluminum oxide to increase its emissivity and thereby allow higher power inputs without overheating of the induction coil. For the aluminum oxide-coated coil, power inputs of 30 and 35 kilowatts were obtained in two runs, as compared with 18 to 22 kilowatts for the uncoated coil. The higher power inputs gave cadmium evaporation rates of 69.5 and 86.5 kilograms per hour, as compared with evaporation rates of 25 to 43 kilograms per hour for the induction coil when its surface was not oxidized.

c. Calorimetry - A second series of combustions of aluminum in fluorine has been completed. The preliminary value for the heat of formation of aluminum fluoride, -359 kcal/mole, agrees reasonably well with the literature value of -357 kcal/mole.

Techniques for the combustion of niobium and tantalum in fluorine have been developed, and samples for the calorimetric experiments are being prepared.

3. General Chemistry and Chemical Engineering

a. Conversion of Uranium Hexafluoride to Uranium Dioxide

(1) Simultaneous Reaction Studies: Preparation of High-Density Particles - Development studies continued on a fluid-bed method for preparing high density, spheroidal uranium dioxide particles directly from uranium hexafluoride. The scheme involves simultaneous reaction of steam and hydrogen with the hexafluoride. By using higher temperatures, 700°C as compared to 650°C, a further increase in particle density to 9.7 g/cc, (88.5 percent of theoretical density) from a previous high of 9.5 g/cc has been achieved.

(2) Fluid-Bed Calcination Studies in Small-Diameter Columns - These studies have been discontinued in favor of other studies of a more general interest.

IV. PLUTONIUM RECYCLE

Plutonium Recycle Study - To obtain approximate values of the plutonium feed enrichment necessary to maintain an EBWR type reactor critical for various degrees of burnup, several CYCLE problems were run assuming a uniform linear mode of operation and utilizing three group cross sections from a previous EBWR Core 2 analysis. The necessary feed enrichment varied from 2.2% to 6.8% depending upon the reactor power and the specified fuel burnup. Enrichments in this range can be obtained in fast reactor blanket elements.

These calculations will be repeated using a 4-group cross section set obtained by use of the MUFT and SOFOCATE codes. The break points between groups are at 0.183 Mev, 1.44 ev, and 0.625 ev. An analysis of the MTR plutonium loaded core using cross sections obtained from MUFT-SOFOCATE results in a calculated excess reactivity 2% less than the experimental value. To produce agreement it is necessary to increase the fission cross section of Pu^{239} obtained from SOFOCATE by 3%; such an increase is appropriate and is confirmed by other studies (CEND-146).

For a typical system ($V_{H_2O}/V_{UO_2} = 1$; $V_{Zr}/V_{UO_2} = 0.24$; rod diameter = 1.27 cm), the range of variation of group cross sections for the various materials has been determined for a wide range of Pu^{239} and Pu^{240} enrichments (1.5%→7.0% Pu^{239}; 0.1→1.0% Pu^{240}). The microscopic cross sections of U^{238} and Pu^{239} were approximately constant over the entire range, but σ_a^{40} varied considerably in groups 2 and 3 as did the transfer cross section, $\Sigma_{3\to4}$ and Σ_{tr} in group 3 for the homogenized material.

V. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Facility Design

The final report by the United Nuclear Corporation on its Preliminary Feasibility and Cost Study on AARR, UNC-5024, has been received. The report presents the current design of the AARR facility by means of drawings and accompanying description material. A detailed construction schedule shows that if Title I design is initiated in November, 1962, normal full-power operation of the plant can be achieved in August, 1966. A detailed cost estimate is also included. Copies of the report have been forwarded to the AEC, together with a revised Schedule 44 Construction Project Data Sheet.

2. Research and Development

a. Core Physics Calculations - Analyses show that the use of hafnium shields to line the flux trap and surround the core will reduce the peak power and keep the power distribution nearly constant with burnup. Unfortunately the shields also reduce the peak thermal flux and reactivity severely; thus it appears that such shields will be useful largely for reduction of local peaks at corners. Graded fuel concentration will have to be used for the over-all power peak reduction.

An analysis of the response to reactivity excursions shows that at ramp rates up to 10 cents per second incipient boiling does not take place until at least three seconds have elapsed; this is ample time for the control system to respond. The reactor period under these conditions is of the order of 10 sec, so that indication of an excursion would be most reliably obtained from hot channel exit temperature indicators or flux level monitors.

b. Critical Experiment - Because of budget restrictions, a decision has been made to delay ordering of fuel for the AARR critical experiment until the latter half of FY 1963, so that the major portion of the fuel will be delivered after the beginning of FY 1964. It is believed that the resulting delay in starting experimentation can be made up by a more intensive effort during a shorter experimental period, so that the schedule for specifying and ordering the first core for the reactor will not be affected.

The intervening period will be utilized to prepare a cell for the experiment, and to develop less expensive fuel elements for the critical experiments.

c. Core Hydraulic Tests - Hydraulic flow tests were conducted on a stainless steel dummy fuel subassembly with no end boxes attached. The flow was varied from about 23 ft/sec (7 m/sec) to 43 ft/sec (13 m/sec) through the subassembly while the coolant temperature ranged from 90°F (32°C) to 210°F (99°C). Pressure drop data and motion pictures were obtained.

Analysis of the data indicates that pressure drops under such flow conditions can be predicted by customary relationships; experimental and calculated values agreed within ± 4%. From the measured data a pressure drop of 80 psi (5.4 atm) was obtained for the subassembly without end boxes, in contrast to the 70-psi (4.8 atm) pressure loss given in ANL-6451* for an aluminum test section. It was estimated that the "keys" used to fasten the plates together accounted for ~15% of the total pressure drop.

Stroboscopic examination of the stainless steel section under flow conditions showed no vibration or detectable deformation of the subassembly during test, in contrast with earlier tests on an aluminum subassembly (see Progress Report for December, 1961, ANL-6485). Post-test measurements showed no permanent deformations. The design appears to be adequate, at least under isothermal conditions.

B. Direct Conversion Studies

The description of experiments in which heat is converted directly to radio frequency power in a plasma diode was previously described in ANL-6525 (Progress Report for February, 1962). The diode consisted of a tantalum emitter, a copper collector and guard ring and was filled with potassium vapor (plasma) to neutralize the space charge and to provide a low work function on the collector and guard ring. A new series of experiments were performed with this apparatus in which cesium vapor was substituted for the potassium with an emitter-collector distance of 2.1 mm.

Emitter temperature, cesium pressure, and the voltage between emitter and collector could be varied. A series of experiments were performed keeping the cesium pressure and emitter temperature constant, and varying the applied voltage between the collector and grounded emitter from about +5 to -5 volts. The voltage-current characteristic curves of the cell were traced by an X-Y recorder.

The difference between the emitter and collector work functions could be determined from the observations made at the knee of the characteristic curve and was found to be between 2.7 to 2.8 volts. Assuming an emitter work function of 4.2 volts, this would indicate the work function of the collector was 1.5 volts, which is slightly lower than noted in most published reports.

Additional experiments were performed keeping the cesium pressure constant, but increasing the emitter temperature (and current). The emitter temperature was varied from about 1600°K to 2400°K. Subsequently the cesium pressure was increased from about 10^{-5} mm Hg to 10^{-2} mm Hg and

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

the series of measurements were repeated; r-f oscillations were observed and their frequencies, rms voltages and power maxima determined.

As in the case with the potassium, the r-f frequency decreased with decreasing d-c voltage and increased with electron emission (current). The frequency, however, sometimes varied in an irregular fashion. The shape of the waves would vary from nearly sinusoidal to rather complicated forms. At the higher vapor pressures, the frequency of the output could not be determined, but an r-f voltage was recorded on the rms voltmeter. The ratios of the d-c to r-f power maxima did not exceed 12%, considerably less than the 25% ratio observed with the potassium cell.

Oscillations were also observed when the collector was biased from -5 to -10 volts with respect to the emitter, i.e., more negative than the open circuit voltage of approximately -3.7 volts. The oscillations appeared to be of the order of 300 to 400 kc on which other frequencies between 50 to 70 kc were superimposed.

VI. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal Oxidation and Ignition Studies

Studies of the oxidation and ignition of plutonium are continuing (see Progress Report, April 1962, ANL-6565, page 51). Shakedown runs with uranium and preliminary runs with plutonium have been performed in a cell on the microscope stage. The strain-gauge pressure measurement of the oxygen consumption and the microscopic observation procedures have been tested. Previous work has shown that oxidation occurs in two distinct stages at 140 and 185°C. Preliminary results from plutonium oxidations at oxygen pressures of 20, 150, and 740 mm indicate a pressure dependence of the second stage rate at 185°C. The rates varied from 8.3 to 21.5 mg/(sq cm)(min) at 20 and 740 mm pressure, respectively. No appreciable effect of pressure was observed in the first stage. Simultaneous observation and oxidation measurements are expected to provide some insight as to the processes occurring.

2. Metal-Water Reaction Studies

Studies of the stainless steel-water reaction by the condenser discharge method are continuing (see Progress Report, April 1962, ANL-6565, page 50). Runs were made with 30- and 60-mil wires in heated water. Runs were performed in pairs in which the only variable was the water temperature: one run in water at 100°C (15 psia) and the other in water at 200°C (225 psia). No difference in the results between runs in water at 100°C and 200°C was found which is in agreement with previous results reported for zirconium. Runs in water at 315°C (1500 psia) are planned for the near future.

Somewhat greater reaction occurred in heated water than in similar runs in room-temperature water. This finding was consistent with previous results with zirconium and uranium. The difference between the extent of reaction in heated water and in room-temperature water, however, was much less in the case of stainless steel. The total extent of reaction at very high initial metal temperatures for stainless steel runs (e.g., 40% reaction at an initial metal temperature of 3500°C, mean particle diameter about 200 μ) was much less than with zirconium or uranium under similar conditions. These two facts indicate that reaction is controlled primarily by a chemical rate law at the metal surface even at very high temperatures. Gaseous diffusion of water vapor through the evolving hydrogen appears to be less important in the stainless steel case than in the uranium and zirconium case.

In-pile studies of metal-water reactions in the TREAT facility are continuing. Two experiments with zirconium -10.65 weight percent uranium alloy plates extended previous data (see Progress Report, May 1962,

ANL-6573, page 52). A total of five experiments have been conducted. At an energy (calculated) of 895 cal/g, 67.2 percent of the plate reacted. At energies below this, and as high as 715 cal/g, the plates had only reacted to the extent of 11.5 percent.

B. Fast Reactor Safety Studies

1. Experimental Meltdown Program

Transient in-pile experiments are being performed in the TREAT reactor in order to study the important mechanisms which produce failure in fast reactor fuel elements and govern the motion of the meltdown products.

a. Meltdown Experiments on Pre-Irradiated EBR-II Elements - The three irradiated EBR-II Mark-I fuel elements which had been remotely assembled in opaque capsules (see Progress Report for June, 1962, ANL-6580) were run in TREAT. Equipment for use in handling the samples at TREAT, and in loading them into the reactor core worked smoothly with a minimum of difficulty. The capsules were received at Argonne and preparations are underway to perform the post-irradiation examination. TREAT irradiation conditions for the samples were as follows:

Sample	Estimated Burnup (at-%)	Reactor Energy Release (Mw-sec)	Maximum Sample Cladding Temperature Recorded (°C)
1	0.5	44	840
2	0.5	50	910
3	0.5	66	1120

Sample burnup is estimated above on the basis of nominal MTR irradiation conditions, corrected by results of relative gamma ray activity of samples.

b. Photographic Meltdown Experiments on Uranium Oxide Fuel Samples - The four samples in Series XXXII for surveying failure characteristics of refractory metal clad ceramic fast reactor fuel (see Progress Report for June, 1962, ANL-6580) were run at TREAT. Two samples have been received at Argonne, but were not examined during the report period. Transient irradiation conditions are given in Table XI.

Preliminary study of the photographs from the series indicates that an unacceptably large amount of clouding occurs in the higher temperature transients (e.g. 5 and 6) due to overheating of the high reflectance paint lining the transparent capsule. The paint is decomposed in the vicinity of the sample by heat transferred from the sample.

Table XI. Transient Irradiation Conditions

Sample No.	Transient	Cladding	Reactor Energy Release (Mw-sec)	Maximum Recorded Cladding Temperature (°C)
1	1	Nb	150	2170
2	2	Nb	164	2380
3	3	Ta	85	1215
3	4	Ta	142	1880
3	5	Ta	204	2300
4	6	Ta	284	Thermocouple broke early in transient

2. Meltdown Equipment Development

a. Large Sodium Loop - A large sodium loop, designed to be a semipermanent installation at TREAT, is being constructed for exposure of clusters of sample elements in flowing sodium under full EBR-II design flow conditions. Design of several components were completed. These items consist of plugging indicator, cold trap, and fission product gas trap. Pressure vessel modifications were also made.

The power distribution transformers required to supply power to the large sodium loop heaters were received and installed in the reactor building. Work was continued on the instrumentation and control panels.

b. Transparent Facility for Photography of Experiments on Pre-irradiated Samples - The design of the transparent facility for use in the photographic TREAT experiments is continuing on the basis that the irradiated sample will be assembled in its refractory metal inner subassembly and steel outer subassembly at the DuPage site and will be also disassembled here after exposure in the TREAT reactor.

3. Sodium Vapor Pressure Measurement

Difficulties were encountered again in the operation of the sodium vapor pressure experiment. This time, the electrical conductivity device for detecting sodium boiling began to give erratic results as high temperatures were approached. Consequently, two alternate techniques were incorporated into the design for detection of fluid boiling. The first is the monitoring of the break in electrical continuity between thermocouple well and crucible occurring when the sodium boils away. The second consists of recording the temperature of the sodium in the crucible. Utilizing these two observations, it has been possible to see the simultaneous ending of the constant temperature boiling range in the "break" of the electrical circuit. Using this technique the pressure range from 7.5 to 11 atm was rechecked. A higher measurement at 13 atm was terminated when a small hole opened in the side of the crucible permitting sodium to leak out.

VII. PUBLICATIONS

Papers

REJECTION OF GAMMA BACKGROUND PULSES IN HORNYAK BUTTONS

A. DeVolpi and K. G. Porges

IRE Trans. Nuclear Sci. 9, No. 3, pp. 320-326 (June, 1962)

EXPERIMENTS WITH NEUTRON SCINTILLATORS MADE OF LITHIUM FLUORIDE, ZINC SULFIDE AND LUCITE

F. Helm

Rev. Sci. Instruments 33, 687 (1962)(L)

CRITICAL EXPERIMENTS RELATED TO BORAX V

K. E. Plumlee

Symposium Summary for Nuclear Superheat Meeting No. 5, TID-7630, pp. 60-61 (1962)

ELASTIC AND INELASTIC SCATTERING OF FAST NEUTRONS FROM Th^{232}

A. B. Smith

Phys. Rev. 126, 718 (1962)

A REDUNDANCY CHECK FOR ALGOL PROGRAMS

Henry C Thacher, Jr.

Comm. Assoc. Comp. Machinery, Vol. 5, pp. 337-342 (June, 1962)

ALGOL 60, AN ALGEBRAIC COMMON LANGUAGE

Henry C. Thacher, Jr.

EDP in Life Insurance, Proceedings of the Automation Forum Held March 19-21, 1962, Chicago, Illinois; Life Office Management Association, New York, N.Y., pp. 263-265 (1962)

THERMALIZATION AND DIFFUSION PARAMETERS OF NEUTRONS IN ZIRCONIUM HYDRIDE

J. W Meadows and J. F. Whalen

Nuclear Sci and Eng 13, 230-236 (1962)

PARTITION OF SOLUTES BETWEEN LIQUID METALS. I. THE ALUMINUM-CADMIUM SYSTEM

F. A. Cafasso, H. M Feder, and I. Johnson

J. Phys. Chem 66, 1028 (1962)

The following papers were presented at the Seminar on Physics of Fast and Intermediate Reactors, Vienna, August 3-11, 1961, and have now been published in Proceedings, Volume I, 1962 International Atomic Energy Agency, Vienna:

CALCULATION OF FAST-NEUTRON REACTION CROSS-SECTIONS
P. A. Moldauer, pp. 171-177

AN ACCURATE TREATMENT OF RESONANCE SCATTERING IN LIGHT ELEMENTS IN FAST REACTORS
H. Hummel and A. Rago, pp. 231-250

DESIGN AND CONSTRUCTION OF THE ARGONNE FAST CRITICAL FACILITY (ZPR-VI)
W. Y. Kato and L. R. Dates, pp. 295-312

INTEGRAL PHYSICS DATA FOR FAST-REACTOR DESIGN
W. B. Loewenstein and D. Meneghetti, pp. 415-434

RECENT ADVANCES AND PROBLEMS IN THEORETICAL ANALYSES OF ZPR-III FAST CRITICAL ASSEMBLIES
D. Meneghetti, pp. 457-487

A CRITICAL SUMMARY OF MICROSCOPIC FAST-NEUTRON INTERACTIONS WITH REACTOR STRUCTURAL, FISSILE AND FERTILE MATERIALS
A. B. Smith, pp. 29-54

EXPERIMENTAL STUDIES OF FISSION PROPERTIES UTILIZED IN REACTOR DESIGN
D. Butler, S. Cox, J. Meadows, J. Roberts, A. Smith and J. Whalen, pp. 125-137

ANL Reports

ANL-6362 THE DISTILLATION OF URANIUM HEXAFLUORIDE AND BROMINE PENTAFLUORIDE IN A 0.5-INCH-DIAMETER PACKED COLUMN
Richard O. Ivins

ANL-6458 KINETICS OF TREAT USED AS A TEST REACTOR
C. E. Dickerman, R. D. Johnson, and J. Gasidlo

ANL-6515 SECOND SYMPOSIUM ON PHYSICS AND NON-DESTRUCTIVE TESTING. Held at Argonne National Laboratory October 3, 4, 5, 1961

ANL-6519 CRYSTALLOGRAPHIC D-SPACE COMPUTER PROGRAM
M. H. Mueller, E. F. H. Meyer, and S. H. Simonsen

ANL-6539 IDAHO DIVISION SUMMARY REPORT, April through September 1961

ANL-6546 CHLORINATION OF URANIUM AND FISSION PRODUCT OXIDES IN MOLTEN HALIDE MEDIA
J. P. LaPlante, D. A. Wenz, and R. K. Steunenberg

DIFFERENTIAL SCATTERING CROSS SECTIONS FOR SLOW NEUTRONS IN D_2O. Reactor Physics Constants Center Newsletter No. 4
H. Greenspan and I. G. Baksys

DIFFERENTIAL SCATTERING CROSS SECTIONS FOR SLOW NEUTRONS IN Be AND BeO. Reactor Physics Constants Center Newsletter No. 5
H. Greenspan and I. G. Baksys