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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

February 1962

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Issued March 15, 1962

Operated by The University of Chicago
under
Contract W-31-109-eng-38

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FOREWORD

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

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

The last six reports issued
in this series are:

August 1961	ANL-6409
September 1961	ANL-6433
October 1961	ANL-6454
November 1961	ANL-6473
December 1961	ANL-6485
January 1962	ANL-6509

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I. WATER COOLED REACTORS

A. EBWR

1. Operation

EBWR is now fully loaded with 147 fuel elements and a neutron source. Of the 28 spikes in the ring, 12 have one boron-stainless strip each, and 16 have two strips each (an average of 1.57 strips per spike). Four additional spikes, each containing three boron stainless steel strips, plus an enriched thin fuel element, were placed in the periphery of the core and are not considered in determining the average strips per spike. This core arrangement now has approximately 4.6% more reactivity than the previous partial loading with 20 spikes. There should also be an additional gain in reactivity of approximately 1.5% due to the new control rods with Zircaloy-2 followers. The previous control rods were equipped with fueled stainless steel followers.

The current loading requires approximately 3.1 gm of H_3BO_3 per gallon of water to meet an eight-rod shutdown requirement. Extrapolation of bank criticality data indicates that the reactor will also be shutdown with nine rods fully inserted and no H_3BO_3 present.

2. Examination of Boron Strips

Plans are being made for destructive examination of an irradiated boron-stainless steel strip. This examination will include determination of the physical, chemical, and metallurgical properties of the strip.

3. Detection of Fuel Growth

A study was made to determine the feasibility of detecting growth in EBWR fuel plates before failure occurred. Such growth could occur from the high operating temperatures expected in some of the fuel plates during 100-Mw operation.

The results of this study are summarized as follows:

a. Case I - Limited Growth - In this case the growth in the fuel plate(s) of an element will result in a corresponding reduction in thickness of the adjoining water channel and in the volume integral of thermal flux. The latter may be measured directly with the aid of a small detector such as a $\frac{1}{8}$ -in. diameter ion chamber. For 40% growth in two adjacent fuel plates, there would be a reduction of approximately 20% in one reading of the detector. With the use of such small detectors, the power generated in each water channel can be determined.

b. Case II - Extensive Growth - With extensive growth in the fuel plates, there would be a corresponding decrease in the volume integral of thermal flux and in the volume integral of power density, or reactor power output. Such growths will be the near equivalent of voids in the core, and loss of reactivity will result. The effect of a uniform growth of 20% in the fuel would be to reduce the core reactivity by approximately 2%. This could easily be detected.

4. EBWR Hydrodynamics

Several new riser configurations were proposed as a method of reducing the carryunder of steam in the downcomers in the reactor vessel expected at 100-Mw operation. Two of the proposed designs are being studied in detail. The first design uses a tapered riser which restricts the core flow and also increases the downcomer area at the top of the riser. (Carryunder decreases as the downcomer velocity decreases.) The alternate riser design consists of two concentric shells. In this case, a straight inner riser is placed over the "spike" fuel elements which are in the central core region bordered by the peripheral control rod crosses. The straight inner riser offers relatively small restriction to flow in contrast to the tapered outer riser which severely restricts the flow from the peripheral (low void) region of the core. All of the RECHOP problems have been completed and a recommendation for selection of riser design will be made.

B. BORAX V

1. Operation

Criticality was achieved at 12:00 p.m. February 9, 1962 with a minimum size core of 14 boiling fuel assemblies containing 49 fuel rods, each of 4.95% enrichment, and a total of 11.2 kilograms of U^{235} . The reactor was critical with the No. 1 (central) rod at 23.4 in. and the remaining rods withdrawn (at 29.0 in.). The reactor was later made critical with the effective rods, 1, 2, 4, 6, and 8 at 23.63 in. and rods 3, 5, 7, and 9 (located in the reflector well outside the core) at 29.0 in. The location of the control rods and instruments, and the initial critical loading are shown in Figure 1.

Channels I, IV, and V were counting channels using fission chambers as detectors. Channel II was a counting channel using a BF_3 detector. Channels III, Period I and II, and Safety I and II used compensated ion chambers.

Following criticality two additional assemblies were loaded to make a square, 16-assembly core and a limited check of the reactivity worth of control rod No. 1 at about the 16-in. position was made. The reactor water temperature was raised 23°F by means of the electric preheat system and the temperature coefficient of reactivity of the 16-assembly core was measured to be approximately zero.

Figure 1

Minimum Critical Loading, BORAX V Boiling Core

Then the Channel III thimble was relocated to the downcomer area just outside the south corner of the core structure and Period I and Safety I chambers were moved to the same location outside the east corner. The source was moved to thimble D.

Loading of boron-stainless steel poison rods into the reactor was started as a preliminary to the buildup to a full-size, 60-assembly core. The poison rods are being loaded in a configuration to flatten the over-all and local radial flux distribution, to adjust the final excess reactivity in the full size core, and to reduce the worth of the central control rod to assure that the reactor cannot be made critical by the withdrawal of a single control rod.

Loading of fuel rods and poison rods into assemblies, and assemblies into the reactor, is continuing. At month's end, 38 fuel assemblies containing 24 poison rods had been loaded. Considerable operational delay was caused by malfunctions in the loading counter circuits, linear amplifier circuit and particularly by spurious scrams on a period trip circuit. Also a control rod drive motor burned out and was replaced.

2. Installation and Modification of Reactor and Components

The control circuits for the two-speed, intermediate-control-rod-gang drives were modified to provide for insertion of the control rods at the high speed. The excessive insertion time (96 min. for all rods) was reduced by 48 minutes. The withdrawal speeds remain unchanged and no additional hazards are involved in this change.

New control-rod-drop timing switches and associated wiring, relays, etc., have been installed at the 4-in. position, just above the dashpot, to permit timing the free fall to the dashpot. An additional manual scram switch was installed on the east end of the sampling station convenient to the reactor pit.

Installation of thermal insulation on the additional stripheaters on the auxiliary water system was completed. All materials are on hand and modification has started on the differential pressure control system and flow-indicating system for the auxiliary water pump and control-rod-drive seals. The existing control-rod-drive sealwater valve is being converted for the required pressure-reducing service.

Work is nearly complete on installation of the low steam flow transmitter and the pressure-to-current transducer necessary for the turbine warmup phase of operation.

The fission product monitor for superheated steam is essentially complete. Some repiping was done on the reactor water fission product monitor to improve its operation. The suspended solids totalizer has been adjusted and calibrated to an accuracy of 0.2% and is now operating. Fabrication and installation of the guide anchors for the superheater drain line were completed.

To control the feedwater temperature at zero power conditions, stainless steel coils for cooling the raw water were installed in the feedwater storage tank.

3. Procurement and Fabrication

a. Superheater Fuel - With the exception of 25 FCE plates which showed rust spots and are still being corrosion tested in room temperature water, inspection is complete on 380 central superheater fuel plates.

Four-hundred and sixty peripheral superheater plates have been received. Out of 89 of these plates inspected this month, 1 was rejected, and 4 required corrosion testing because of the appearance of rust colored stains on the face cladding.

The rust colored stains which appear on the surface of some plates are caused by corrosion of foreign particles which were embedded in the face cladding during the rolling operation. All evidence indicates that these particles are ferritic. It is difficult to find these particles during the visual examination because, as a rule, they are under 0.010 in. in length. However, after the plates have been in water for $\frac{1}{2}$ hour the presence of such particles becomes apparent due to the formation of the rust colored corrosion product. In some cases the rust stain is $\frac{1}{4}$ in. in diameter. Microscopic examination of the area reveals a black stain surrounded by the reddish-brown stain. These colors correspond to the colors of two oxides of iron, namely FeO and Fe₃O₄.

Attempts were made to remove the embedded particles by scraping. After scraping, the plate was placed in a tank of distilled water for 16 hours to check the effectiveness of the scraping. Rust stains reappeared on approximately 50% of the scraped areas. However, in all cases the black oxide was gone leaving a pit in plate surface. The bottom of the pit was a metallic silver in color with a faint trace of rust. After cleaning and retesting no further evidence of corrosion appeared. It was then decided to remove the particles by promoting corrosion of the iron. This is accomplished by leaving the plate in water until the reaction is complete (Fe-FeO-Fe₃O₄). The reaction probably follows these basic steps. The depth of the pit is then measured using a metallurgical microscope. If pits deeper than 0.002 in. are observed the plate is rejected. Nine of the 21 type FCE plates which had particles embedded in the cladding have been tested in the above manner. All nine were acceptable.

Facilities for vacuum brazing of the plates into elements have been increased by the installation of a second 1375°C glow bar furnace. It is now possible to braze five elements simultaneously. Two such groups were vacuum brazed during the past month. All plates used contained depleted uranium. Of the 10 elements made, eight appeared to be brazed satisfactorily. One element was slightly discolored on one end because of a small leak in the evacuation can. The other element had adhered to the Hastelloy-C cover plate of the brazing fixture because coating of aluminum oxide was not enough to prevent such bonding. Development work is being continued to perfect the process.

b. Reactor Components - Fabrication of the integral and closure pieces for the new control rods is complete. Modification of the aluminum follower and cruciform stainless steel extension to strengthen the mechanical joint between the two is proceeding. All spot-welding slugs have been made and forming of the 304L stainless steel cladding has started.

Eight 17-4 PH stainless steel, control rod extension shafts have been completed and inspected. Three additional shafts must be stripped and replated.

During the reactor loading operation, 16 defective boiling fuel rods and 3 defective boiling fuel assembly boxes were discovered. Most of these can be repaired.

c. Experimental Components - Fabrication of the latches for the exit flowmeters was completed and assembly of the instrumented boiling fuel assembly continued. Development work on the pressure seal section of the instrument leads is also continuing. The only apparent development problem remaining is that of making the final braze between instrument leads and the pressure seal section.

Specifications have been prepared and bids requested for two types of miniature fission detectors for flux mapping operations at-power. One detector type is a fission heating device. Both types are to be used for obtaining total and epithermal fission rate measurements in U^{235} . Totally enclosed cadmium liners on the detectors are proposed as the "filtering" method.

It is anticipated that the fission heating device may permit both measurements to be made in one assembly. Limitations of space indicate that two separate ion chambers will be needed to obtain the required information on total and epithermal fission rate.

Fabrication of the movable steam probe is continuing. Modifications are being made to permit this device to be used as a source drive during high temperature zero power operation. Components have been procured and fabrication has started on the drag-disc-type downcomer flowmeter and on 50 aluminum, axial flux wire holders to be used in the coolant channels of the boiling fuel assembly for zero power flux mapping.

Procurement and construction of the test equipment and electrical control panel for the H_2-O_2 recombiner test rig is well advanced. Samples of woven-wire-mesh demister material have been received. This material, made of nichrome and Inconel, will be used as a base for the platinum catalyst coating. A gas transfer apparatus to be used with the gas analysis equipment on both the recombiner test rig and the reactor is being fabricated.

Difficulties have been encountered in fabricating a leaktight, X-8001 aluminum cladding for the Boral section of the oscillating rod. The use of Zircaloy-2 as a cladding material is now being investigated. The components for the in-vessel drive shaft and bearing section of the oscillating rod were completed.

Fabrication and procurement for the root-mean-square noise analysis equipment is well advanced.

4. Design

Detailed design was completed on the equipment to test the performance of a platinum catalyst H_2 - O_2 recombiner in steam at 600 psig saturated conditions. Detailed design continued on the step function generator equipment and the fuel handling coffin and "head-on" fuel handling tool. The superheater fuel element grapppler has been redesigned to make it fail-safe in the closed position on failure of dc solenoid power.

The storage racks and storage pit arrangements for both boiling and superheater fuel are being redesigned to insure subcriticality without the use of cadmium.

5. Development and Testing

a. Plant Tests - After three weeks of operation, the boron addition system was test-operated to assure flow of the heated, supersaturated (at room temperature) boric acid solution. The final boric acid tanks-to-reactor solenoid trip valve was removed and a temporary flange and hose-to-sump were installed. With the tank temperature at $210^\circ F$, the boron addition switch was operated and all trip valves operated properly in sequence. There was a two-minute delay before flow was initiated. The flow of clear solution was preceded by rusty crud and mud. The heated boron tank drain line and the upper boron tank level gage glass were found to be plugged. The plugged drain line was cleared by raising the tank pressure to 30 psig. The plugging of the sight glass is apparently due to local overheating and boiling. These defects are being investigated. Meanwhile, the boron addition system is operational.

As a result of the difficulties with the supersaturated, heated boric acid solution, an investigation is being made of using a boron compound which will contain sufficient boron in a subsaturated solution at room temperature. At present "Polybor," sodium octoborate, appears to be the most promising compound.

b. Control Rods - Development work on the cladding-to-slug, spot-welding technique was completed. Special welding fixtures and offset water-cooled electrodes was designed and fabricated to assure good spot welds with no expulsion. A prototype cruciform poison section was assembled and welded to check production techniques.

A dummy control rod of reduced width has been made. This rod will be subjected to repeated scrams in the EBWR control rod drive test rig to prove the design of the revised mechanical joint between the aluminum follower and the stainless steel cruciform extension.

c. In-Core Instrumentation - The reliability and the useful upper temperature limit of the high temperature, fuel rod thermocouples must be increased. For this purpose a literature search is being made to obtain the most recent information concerning the properties of high purity, sintered metal oxides. This information is important because of the low insulation resistance of arc-fused materials at temperatures approaching 4000°F. Arc-fusion introduces impurities which may tend to make the insulator more of a semi-conductor.

Another possible improvement is to change the sheath material of the above assembly from tantalum to tungsten-rhenium alloy. This would provide a more ductile material and lessen the chances of failure due to the brittle character of the former material after temperature cycling.

d. Corrosion of Structural Materials in Superheated Steam - Testing of stainless steels in oxygenated superheated steam at 650°C and 600 psi continues (see Progress Report, December, 1961, ANL-6485). Types 309 and 310, as well as 406, appear to be superior to 304, 316, and 347. Of the last three materials, 304 is perhaps the least resistant. However, it still appears that corrosion rates are decreasing with time. It is also indicated that the type of surface treatment has an important influence on corrosion of these steels in the present environment.

e. Exponential Experiment - The fuel rod assembly for the BORAX V exponential experiment in the TREAT shield was loaded up to a 349-rod, cadmium-covered cylinder. A number of axial and radial flux traverses were run on the assembly. The detailed fine structure of the radial flux was determined across a simulated center 49-rod fuel assembly, (a) with no voids, (b) with voids, (c) with a water hole in the center, (d) with boron-stainless steel, and with stainless steel rods in the corners. Some difficulty was experienced initially in obtaining satisfactory activation and β discrimination on the 3.4 w/o U-Zr flux wires. However, the use of $\frac{3}{8}$ in.-long pieces of chopped wire and a temporary modification of the flux wire counter has yielded good results.

A variation of moderator void content was simulated with closed polyethylene tubes and this was related to the cadmium ratio. Typical values obtained thus far are as follows:

<u>% Void</u>	<u>Cd Ratio</u>
0	6.17
4	6.03
8	5.62
16	5.56*

* In this case, the void representation was changed in such a manner that the flux wire was largely in the void.

Further measurements are being made in order to determine the feasibility of this method of void measurement for later use in BORAX V.

II. LIQUID METAL COOLED REACTORS

A. General Research and Development

1. Evaluation of Carbide and Oxide Ceramic Fuels for Large Fast Breeder Reactors

An attempt is being made to assess the ultimate potential of uranium oxide and carbide fuels in terms of achieving minimal fuel cycle costs for large fast breeder reactors. Effort is directed toward a parametric heat transfer analysis of fast breeder cores using these fuels. This heat transfer data will be used in conjunction with the CYCLE code to calculate fuel cycle costs for typical breeder cores using each type of fuel.

The reference problems shown in Table I have been selected for analysis using the CYCLE code.

Table I. Problems for Cycle Analysis*

Problem No.	Core Volume (liters)	Fuel	Rod Diameter (in.)	Coolant Velocity (ft/sec)	Efficiency (%)
179.40001	6080	Oxide	0.25	42	41.1
179.40002	601	Carbide	0.25	109	40.8
179.40003	1404	Carbide	0.25	30	29.7
179.40004	2676	Carbide	0.50	30	40.5
179.40005	2676	Oxide	0.125	30	41.1

*All codes give an output of 1338 Mwt; the fuel cycle analyzed is a U^{238} - Pu^{239} cycle.

Some exploratory work has been done on improving cost numbers used in the cost code calculations. The following relationships, based mainly on data reported in Y-1368,** have been used for the fabrication costs of the oxide and carbide fuels:

$$\text{Oxide: } \text{Fabrication Cost } (\$/\text{kg}) = 152.3/(d)^{0.6}$$

$$\text{Carbide: } \text{Fabrication Cost } (\$/\text{kg}) = 190.5/(d)^{0.6}$$

**P. E. Stein, et al., "The Influence of Design and Production Parameters on Fabrication Costs of Nuclear Reactor Fuel Elements," Y-1368 (September 6, 1961).

The initial enrichments for the five reference problems are in the range of 8%-10% plutonium. Processing costs of about \$100/kg have been used for conventional aqueous solvent extraction processes for both oxide and carbide fuels.

It is anticipated that work on this study will terminate upon completion of the five reference problems. The first two have been completed and the last three are in progress.

2. Isotopic Power Sources

A study has been started to explore the feasibility of making Pu^{238} and Cm^{242} as useful byproducts in the operation of fast power reactors. Appropriate neutron cross-sections have been estimated and the isotope production equations have been programmed for machine computations.

The machine program for calculating production rates was tested. The input data were based on the design operating levels of the Enrico Fermi Reactor which are:

- (1) Power - 300 Mwt, 100 Mwe
- (2) Average core flux - $5 \times 10^{15} \text{ n}/(\text{cm}^2)(\text{sec})$
- (3) Enrico Fermi spectrum
- (4) 1% burnup in the core
- (5) Average core irradiation time - 50 days
- (6) Specific core power - 550 kw/kg U^{235}
- (7) Critical mass U^{235} - 485 kg

The production of Np^{237} was calculated for two starting materials, U^{235} and U^{238} , in both the core and the blanket. The production in the blanket is negligible and the results are not presented here.

Assuming that the Np^{237} can be sold for a \$500/gm profit after chemical separation, the value of Np^{237} produced is given in Table II as a function of the number of fuel recycling steps.

Table II. Value of Np^{237}

Number of Recycles	Total Production Per Cycle (grams)	Np^{237} mills/kw-hr electric
1 - 4	77.21 (avg)	0.326 (avg)
5	232.1	0.9810
6	262.2	1.108
7	289.0	1.222

Based upon observation it appears that, in general, the value of the neptunium will increase an average of about 0.12 mill/kw-hr electric per cycle less the cost of separating the neptunium during the first 10 to 20 cycles.

The following separations are also of interest:

- (1) Am^{241} from irradiated fuel
- (2) Cm^{242} from irradiated fuel
- (3) Pu^{238} from Np
- (4) Cm^{242} from Am

The value of these isotopes as well as reliable costs for separating the end product must be determined to evaluate fully the worth of the production method.

3. ZPR-III

The experimental program with Assembly 35, the mockup of the Enrico Fermi reactor core B, has been concluded. This study, under way for about six months on ZPR-III, has resulted in an extensive survey of the properties of a 600-liter, cermet-fueled, sodium-cooled core. With a cermet fuel element it is necessary to use highly enriched uranium, thus sacrificing the breeding characteristics of the core. It was further found necessary to replace some of the inner U^{238} blanket with a more reactive reflector in order to achieve criticality within the dimensions to which the Fermi reactor vessel is limited. Although this results in some further sacrifice of breeding, the replacement produced such an improvement in reactivity that it was felt advantageous to experiment with a number of blanket materials with a view towards optimization of this feature. A nickel reflector was used for the bulk of the Fermi experiments, but a survey of other possible materials indicated that either a beryllium oxide or a nickel oxide reflector would offer a substantial increase in reactivity. Preliminary experiments indicated that the beryllium oxide was subject to experimental uncertainties in ZPR-III because the proximity of beryllium to fuel (which cannot be varied arbitrarily) has a considerable effect on the worth measurements. Nickel oxide was therefore chosen as the reflector material for the final Fermi studies.

a. Rossi Alpha Measurement - Before beginning the change from a nickel to a nickel oxide reflector, it was desirable to do a Rossi- α measurement. Trouble was encountered with the Rossi- α electronic equipment, however, and reliable measurements were not made until part of the reflector replacement was accomplished. Therefore, the Rossi-alpha experiment was run when the Ni-to-NiO exchange was one third completed. A value of $\alpha = -1.60 \times 10^{-4}$ was found for delayed critical. With $\beta = 0.65\%$ the lifetime is then $\ell = 4.1 \times 10^{-7}$ seconds.

b. Construction of NiO Reflector - In order to mock up a NiO reflector, it was necessary to provide the oxygen content with plates of Al_2O_3 and to represent sodium with aluminum. With the composition arrived at, the thickness required for criticality with the 600-liter core was estimated to be $\frac{2}{3}$ that of the nickel composition. Hence, in the reflector drawers the NiO composition was placed in the eleven $\frac{1}{8}$ -in. columns adjacent to the core and the fine blanket composition (40 v/o U^{238}) was placed in the outer five $\frac{1}{8}$ -in. columns. The composition of the NiO reflector is given in Table III. An average radial thickness of 1.68 in. was required for criticality.

Table III. Composition of "NiO" Reflector

Material	Composition (g/cc)
Ni	2.69
Al	0.71
O	0.63
Steel	0.63

c. Recalibration of Control Rods - Before making worth measurements in this NiO-reflected core, a check was made on the calibration of control rods to determine any change induced by the change of reflectors. Within the accuracy of period measurements, no difference was found in the rod calibration.

d. Reactivity Coefficients in NiO Reflector - Because of the approximations involved in the simulated "NiO" reflector, measurements were made of the worths of materials in this reflector. The results obtained are listed in Table IV and compared to the measurements in the Ni reflector.

Table IV. Reactivity Coefficients in
NiO and Ni Reflectors

Material	Worth in NiO Reflector (Ih/kg)	Worth in Ni Reflector (Ih/kg)
Ni	1.8	1.6
O	6.7	7.0
Steel	1.6	1.5
Na	5.8	7.8
Al	3.9	

e. Worth of Enriched U Columns vs. Radius - The worth of enriched uranium columns at various radii corresponds to fuel subassembly worths in the Fermi Core and is sensitive to reflector composition and thickness. Measurements at five radial locations made in the Ni-reflected core of the worth of 93% enriched U per column were repeated in the NiO-reflected core. The comparison is given in Table V.

Table V. Enriched U Worth/Column vs. Radius in NiO and Ni Reflected Core

Radius (inches)	Worth in NiO-Reflected Core (Ih/kg - column)	Worth in Ni-Reflected Core (Ih/kg - column)
0.0	62.3	61.8
4.4	57.0	56.6
8.7	46.3	45.2
13.04	31.4	30.3
17.5	18.1	17.1

f. Sodium Worth/Column vs. Radius - The worth of sodium as a function of radius in the Fermi Reactor is required by APDA for kinetics calculations. A repetition was therefore made of part of the measurements carried out in the Ni-reflected core. Table VI compares the Na worth per column in the two cases.

Table VI. Worth of Na/Column vs. Radius in NiO and Ni Reflected Cores

Radius (inches)	Na Worth in NiO-Reflected Core (Ih/kg - column)	Na Worth in Ni-Reflected Core (Ih/kg - column)
6.5	17.1	17.4
13.1	13.0	13.5
17.7	8.8	9.4

g. Worth of B¹⁰ in Fermi Safety Rod Channel - There was concern that the change from Ni to NiO reflector may have altered the worth of B¹⁰ in the radial region of the Fermi safety rods. Remeasurement of the worth of B¹⁰ in a 16-in. column at the average rod position indicated no significant change.

h. Foil Irradiations - As part of an investigation of the variation of the fission- and capture-rate with radius through the NiO reflector, foils of enriched and natural uranium were irradiated for 40 watt-hours and analyzed radiochemically. The reduction of these data has not yet been completed.

i. Radial Fission Traverses - The fission rates of enriched and natural uranium as a function of radius at the assembly midplane were also obtained by means of traverses with fission chambers. The results are reduced to count rates relative to rates at the core center and given in Table VII with the corresponding results in the Ni-reflected core. The reflector-core boundary in this table occurs 18.5 in. from the core center. The fission rate pattern for enriched U through the Ni and NiO reflectors is plotted in Figure 2.

Table VII. Radial Fission Traverses in NiO and Ni Reflected Cores

Radius (in.)	Relative Fission Rates (%) With NiO Reflected Core		Relative Fission Rates (%) With Ni Reflected Core	
	93% Enr. U	Nat. U	Enr. U	Nat. U
31.0	5.9	0.1	5.9	0.2
27.2	10.0	1.6	10.0	1.0
22.5	27.0	8.0	28.3	6.0
20.5	41.9	14.9	46.4	13.4
19.3	52.7	22.5	57.5	23.0
18.5	57.0	31.7	59.9	31.7
17.5	59.1	43.8	59.5	41.8
16.2	63.0	50.8	62.2	50.0
14.0	69.2	64.6	68.9	62.4
12.0	76.5	73.8	75.3	70.7
10.0	83.8	81.3	82.4	80.2
8.0	89.5	87.6	89.0	87.5
6.0	94.1	93.0	94.0	93.6
4.0	97.5	96.7	97.5	97.1
2.0	99.8	99.0	100.0	98.9
0.0	100.0	100.0	100.0	100.0

Figure 2

Radial Fission Rate Traverse
of Enriched U through Ni and
NiO Reflectors

j. Uranium Reactivity Coefficient Traverses - Using the same mechanical setup as for the radial traverses of fission chambers, small samples of enriched and natural uranium were moved remotely through the reactor and their worths measured at several radii. Table VIII compares the results with previously obtained worths vs. radii with the Ni-reflected core. A marked change of the worth of depleted U is caused by the change of reflectors, and this is plotted in Figure 3.

Table VIII. Reactivity Coefficients of Enriched and Natural U. vs. Radius at Reactor Midplane for NiO and Ni Reflected Cores

Radius (in.)	Worths (lh/kg) in NiO Reflected Core		Worths (lh/kg) in Ni Reflected Core	
	Enr. U	Nat. U	Enr. U	Nat. U
27.5		0		0.88
23.5	8.4	+1.9	7.9	1.18
20.5	16.3	+1.49	11.2	0.74
18.5	23.0	+1.00	22.0	-0.44
17.5	26.8	+0.89	26.9	-1.18
16.0	35.2	+0.74	34.0	-2.36
14.0	42.3	-0.45	40.2	-2.51
11.0	59.0	-0.74	51.0	-2.51
8.0	69.1	-2.53	67.6	-2.66
5.0	77.9	-3.28	76.7	-2.95
2.0	85.4	-4.17	80.8	-4.72
0.0	88.8	-5.66	83.3	-5.31
-2.0	85.0	-5.81	81.2	-5.61

Figure 3
Radial Traverse of Depleted
U Reactivity Samples through
Ni and NiO Reflectors

Disassembly of this mockup has begun, and during unloading of the fuel, subcritical counts are being taken in the storage vault to determine if any increased multiplication is discernible.

4. ZPR-VI and ZPR-IX

a. Building - The interior walls of Cells No. 4 and No. 5 in Bldg. 315, which will house ZPR-VI and ZPR-IX, are being coated with Carbolite 305, a phenolic resin base paint, in order to decrease the porosity of the concrete walls. Painting of the cells is about 50% complete.

The analytical computations of stresses and strains on the ZPR cells were completed. The calculations were made for a homogeneous and isotropic material with elastic behavior and were based on a model comprised of typical 1-ft wide longitudinal and transverse frame bents in a rectangular box frame. Extremely close agreement was obtained between analytical and experimental strains at the points of maximum positive moments, i.e. at midheight of the transverse frames. However, large discrepancies (as large as 50%) were observed in the strains of the maximum negative moments along the edges. The sources of error in the analytical results are believed to be known and can be reduced by recalculating the results using more realistic dimensions (than those of the centerline) due to the negative moments.

b. Control and Safety Rods - Eleven of the twenty dual-purpose control and safety rod drive mechanisms have been received from the vendor. Wiring of the drive mechanisms is in progress.

Cycle-testing equipment for the dual-purpose rod drive mechanisms has been assembled and testing of the drives will start next month.

All twenty-four insertion safety rod drive mechanisms for both ZPR-VI and ZPR-IX have been received. Wiring of these units will begin after completion of work on the dual-purpose drives.

c. Control Console and Instrumentation - Testing of the instrumentation for ZPR-VI and ZPR-IX is complete except for time constant measurements of some of the trip circuits.

The neutron sensitivity of the BF_3 ion chambers has been checked. The sensitivity for each of the twelve chambers is found to be about 3×10^{-13} amp/unit thermal neutron flux.

5. JUGGERNAUT Critical Experiments

a. Fuel Loading - During the last month the fuel loading has been slowly increased to 3.92 kg of U^{235} , an excess of 520 gm above the cold, clean critical mass. The control rods have been calibrated for various fuel loadings and the fuel worth has been obtained for nine of the eleven fuel additions which have been made.

The measured fuel worth has varied from $+0.0057 (\Delta k/k)/\text{gm}$ of U^{235} to $+0.0125 (\Delta k/k)/\text{gm}$ of U^{235} depending on the location of the added fuel with respect to the shim rods. The best average value for the fuel worth is $+0.0078 \pm 0.0003 (\Delta k/k)/\text{gm}$ of U^{235} . With the beam holes open the excess reactivity of the core is $3.5 \pm 0.15\% \Delta k$.

b. Rod Worths (Assuming $\beta_{\text{eff}} = 0.0075$) - Measurements of the worths of the individual rods have been made at three different fuel loadings. The worths of all the rods are the same within 5% to 10% and equal to $1.0\% \Delta k$. With the beam tubes filled with graphite plugs, the worth of the shim system was measured to be $3.7 \pm 0.4\% \Delta k$ by rod-drop and sub-critical multiplication techniques. Its worth, as determined by the amount of U^{235} it will hold down, is equal to $3.5 \pm 0.2\% \Delta k$ with the beam tubes open.

With all seven shim safeties inserted and the beam holes plugged with graphite, the extrapolated critical loading is 4400 gm. This leads to a calculated safety-system worth of $3.9\% \Delta k$ with a fully loaded core. When the beam holes are unplugged the safety-system worth drops to $3.3\% \Delta k$ (measured by rod-drop technique). The worth of the two safeties (Nos. 2 and 3) which directly shadow beam tubes are each reduced by an amount equal to the worth of the beam tube.

c. Void Coefficient Measurement - In order to simulate voids, teflon plates were inserted between fuel plates in 15 of the 240 core positions, thus replacing 3.1% of the water in the active core. The measured void coefficient was $-0.13\% (\Delta k/k)/1\%$ void compared to a calculated value of $-0.15\% (\Delta k/k)/1\%$ void. The reactivity effect of the teflon was compared to that of air in the central hole when filled with water. Within experimental error, no difference could be detected between teflon and air.

d. Temperature Coefficient Measurement - The JUGGERNAUT has been found to have a constant negative temperature coefficient equal to $-0.014\% (\Delta k/k)/^{\circ}\text{C}$ over the temperature range 20°C to 80°C when heated under equilibrium conditions (5°C increase per hour). When rapidly heated or cooled (10°C to 20°C increase per hour), a prompt temperature coefficient of reactivity equal to $-0.020\% (\Delta k/k)/^{\circ}\text{C}$ is observed. The difference between these two values is attributed to a delayed positive temperature coefficient equal to $+0.006\% (\Delta k/k)/^{\circ}\text{C}$. It has been tentatively concluded that there are two contributions to this delayed coefficient:

(1) A positive coefficient of $+0.008\% (\Delta k/k)/^{\circ}\text{C}$ exhibiting a time delay of 90 min during heating and 30 min during cooling. This effect has been tentatively ascribed to the expansion of the aluminum core containers.

(2) A negative coefficient equal to $-0.002\% (\Delta k/k)/^{\circ}\text{C}$ exhibiting a time delay of several hours. This effect has been tentatively ascribed to graphite heating.

The equilibrium temperature coefficient was calculated to be $-0.020\% (\Delta k/k)/^{\circ}\text{C}$ to $-0.019\% (\Delta k/k)/^{\circ}\text{C}$ as the temperature increases from 20°C to 65°C .

B. EBR-I

1. Fabrication of Core IV Fuel Elements

All of the plutonium fuel slugs have been completed and all available fuel tubes loaded. Additional Zircaloy-2 tubing has been received and inspected. Machining of this tubing is now under way to provide jackets for 100 fuel rods still required.

2. Production of Breeding Gain Specimens

The following technique has been developed for jacketing small plutonium-1.25 w/o aluminum slugs in aluminum cans:

- (1) The plutonium slug is loaded into the aluminum can using a plastic funnel to protect the can from contamination.
- (2) A top cover is placed above the plutonium slug.
- (3) The assembly is pushed through the die to shrink the jacket down through the slug.
- (4) The upper lip of the jacket is spun over the aluminum end cap.
- (5) The end is sealed by a capacitor-discharge weld.
- (6) The assembly is then coined to size in a double-punch coining die.

The technique has been demonstrated on nonradioactive samples using an aluminum alloy as a plutonium slug stand-in. Equipment is being prepared for installation in the gloveboxes.

Two methods are being tested to jacket the 0.150 in. diameter by 0.005 in. thick plutonium disc specimens in zirconium or aluminum foils.

The finished specimens are to be 0.200 in. diameter by 0.010/0.015 in. thick.

(1) The edge welding of a zirconium clad sandwich by a miniature inert gas shielded tungsten arc has been shown feasible. Zirconium foils have been successfully welded down to 0.005 in. but the danger of contamination of the weld and jacket appears to be very great.

(2) The second method consists of positioning the plutonium discs between the aluminum foils between a punch and die assembly. Cover foils are punched and pressure edge-welded in a single stroke operation. It appears that this method might be accomplished with less danger of contamination than method 1, but very careful positioning of the plutonium disc will be required to prevent cutting it as the specimens are punched.

C. EBR-II

1. Reactor Plant

Starting of the primary tank dry heat test was noted in ANL-6509 (Progress Report, January, 1962). Initially, all six electrical immersion heaters operated at approximately 7.5 kw each. Approximately 140 thermocouples were read and recorded once each hour, 24 hours a day. In addition, periodic readings of the primary tank deflections and rotations were made and recorded.

On-off cycling of the immersion heaters was controlled manually. The heaters were shut off whenever maximum permissible temperature gradients within the primary tank structure were reached. The W-5 heater, which is located just a few inches from the primary tank inner vessel wall, was found to cause local "hot-spots" (as expected) and was disconnected temporarily until the vessel reached approximately 200°F. Above this temperature all heaters could be used. The average heating rate for the vessel as a whole was about 1-2°F/hr, and was fairly continuous.

Two of the three additional fans specially installed for this test to provide air circulation in the tank (to simulate conditions prior to filling the tank with sodium) developed mechanical difficulties and the heating was continued without the use of any forced air circulation. The temperature distribution in the primary tank structure found at the maximum temperature reached (about 230°F), while not uniform, did not indicate the existence of large thermal gradients. It thus appears that it will not be necessary to provide forced air circulation when heating the tank prior to filling with sodium.

When the average temperature of the tank reached approximately 230°F, a fuel handling operation was performed transferring a dummy core subassembly from the reactor to the storage basket. The transfer to the storage basket was accomplished satisfactorily. However, when returning the subassembly to the reactor (during the operation where the subassembly is transferred from the transfer arm to the gripper), the subassembly became misaligned with the holddown mechanism funnel. This misalignment caused the bottom of the subassembly to catch on the lip of the funnel as the holddown mechanism was being raised. The upward movement of the funnel continued until the mechanism drive motor stalled, inflicting the damage described below. The dry heat test was then discontinued and the tank cooled down to permit personnel to enter for inspection.

Damage to the holddown mechanism was limited to bent cantilever arms which support the funnel away from the holddown shaft. The gripper jaws were damaged. The end socket fitting on the transfer arm was badly bent. The vertical shaft of the transfer arm was slightly bent. No apparent damage was sustained by the vertical shafts or the drive systems of either the holddown or the gripper.

New supporting arms for the holddown funnel have been fabricated and are being installed. The damaged parts of the gripper jaw assembly have been replaced and the gripper mechanism adjusted and tested. A new end socket fitting for the transfer arm is being fabricated. The vertical shaft of the transfer arm has been straightened and stress-relieved. The latter work required removal of the transfer arm plug from the primary tank and disassembly; reinstallation has not yet been accomplished.

It is estimated that all repairs will have been made, mechanisms checked out, and the tank readied for resumption of the dry heat test early in March.

The cause of the misalignment between the subassembly and the holddown and the resulting interference is not clearly established. Steps presently being taken to preclude repetition of such damage in the future include addition of electrical interlocks to de-energize the holddown mechanism drive motor in the event of interference, and a design change on the transfer arm end socket fitting to assure vertical alignment of the suspended subassembly irrespective of a possible deviation in the horizontal attitude of the transfer arm. Study of other probable causes of the incident and possible further corrective action is continuing.

2. Sodium Boiler Plant

Installation of Evaporator B3-EV702 and the two superheaters is nearly complete. The welding of the stainless steel portion of the secondary sodium piping is about complete. Preparations for the 175 psig pneumatic test and the helium mass spectrometer leak test of the secondary system are underway.

The primary heat exchanger was moved into the reactor plant and lowered into place in the primary tank. Alignment between the heat exchanger and the well casing was satisfactory. Preliminary alignment of the secondary sodium piping external to the building, the reactor building piping penetrations and the heat exchanger nozzles was made. The secondary sodium piping will be temporarily terminated outside the reactor building until the flexible seals, heat exchanger and interconnecting piping are installed.

The pulse echo technique was used to check 6 braze joints on the right and left copper bus bars for an electromagnetic pump for EBR-II. Eighty percent transmission of sound through the braze as compared with the solid copper was considered adequate. The whole of the braze joint is considered good, if this 80% transmission level is kept over 75% of the area of the joint. In February, 6 of these braze joints were inspected; 5 were passed and one was rejected. The reject was rechecked after being taken apart and rebrazed.

This technique of inspection and the standards used for evaluation of these brazes are somewhat arbitrary. This consideration has led to the initiation of a project to correlate the heat transfer across a braze joint with its ability to transmit ultrasound. Six copper specimens with braze joints of varying quality are being made. Measurements of the sound transmission and of the heat transfer across these joints will be compared with the measurements made on a reference specimen, i.e., one with no braze joint. Equipment is now being assembled to heat the specimens to various temperatures, and to make the temperature measurements.

3. Training

The Systems Training Course for staff and supervisors, started about one year ago, was completed in February, 1962. All systems applicable to dry and wet critical operation were covered. Seventy class sessions were held from April 4, 1961 to February 23, 1962. A total of 200 class hours and 400 field session hours were spent in this systems study course.

The technician training program was resumed on February 19 after a recess period dating from January 22 necessitated by the dry heat test.

4. Fuel Reprocessing Facilities

a. Fuel Cycle Facility - Finishing, corrective, and installation work in the Fuel Cycle Facility is being carried out under the Package 4 contract. This work began on January 13, 1962. Photographs taken on this day of the Air Cell, Argon Cell, and Operating Annulus are shown in Figures 4, 5, and 6, respectively.

Figure 4

Inside the Air Cell of the EBR-II Fuel Cycle Facility looking away from the Argon Cell. Conduits rising above the floor near the walls will be used to provide services into the cell. The circular disks at near floor level in center of Air Cell cover ten-foot deep storage pits for fuel subassemblies. The five-ton crane at the top of picture derives its power from the bus bars on the ceiling near the left wall. Below the crane is the bridge for the electro-mechanical manipulator.

Figure 5

Inside Argon Cell of EBR-II Fuel Cycle Facility. On right is the wall of the operating annulus; on left is the wall of the central control room. Groups of five service sleeves (covered with protective plates) are seen at floor level. The sleeves connect with the sub-cell area and will be used to bring services into the Argon Cell. Hinge pins, which will support a remotely replaceable window section, are on the sides of the window openings. Above the window openings are window shutter rails from which will hang shielding shutters to protect the windows from radiation damage. A five-ton crane is in the upper foreground. Manipulator bridges span the work area in the center of the picture.

Figure 6. Part of operating annulus of LBR-II Fuel Cycle Facility looking toward the Argon Cell. The control of fuel reprocessing operations will be carried out from this annulus. The door in the left foreground offers direct access to the Air Cell. The window openings will accommodate five-foot thick shielding windows. The ports above the window openings are provided for mechanical manipulators. The pressure control panel for the Argon Cell atmosphere is on the right.

Work being done under the Package 4 contract includes the following: (1) construction and installation in the subcell of the electro-pneumatic control valve systems (valve cabinets) which will actuate remote refabrication equipment in the process cells, (2) installation of brackets for window shutter drives in the Argon Cell, (3) installation of a pillar jib crane of 5-ton capacity on the roof of the Argon Cell, and (4) installation of work benches and storage cabinets in the machine shop.

Installation of viewing windows in the Fuel Cycle Facility has been started. This work is being carried out by the window fabricator, Pittsburgh Plate Glass Company, through a subcontractor.

Corrective work has started on the dismantling of bearings on the in-cell crane bridges and trolleys. These bearings became contaminated with dust resulting from grinding operations and must, therefore, be cleaned and relubricated.

A purchase order has been issued for the monitoring system for the suspect stack. Specifications for the monitoring system for the off-gas from the melt refining and skull oxidation furnaces are being prepared.

Tests are being made to determine the feasibility of using sounds from the operating manipulators as means of determining whether

the manipulators require servicing. Several audio systems have been tested, but thus far none has proven to be entirely satisfactory.

A second group of four technicians is being hired and training in Fuel Cycle Facility Operations will begin in March. All technicians and staff are now principally engaged in checkout of systems and in noting requirements for completion and correction, and in aiding in Package 4 installations.

Arrangements for procurement of fuel hardware and expendable materials for fuel cycle operation are well under way. Modification of laboratory space to provide facilities for preparation and examination of metallurgical specimens are substantially completed. The laboratory group is preparing for initial analyses of irradiated fission samples. Mass spectrometric analysis of a series of standard samples is substantially complete.

A control system which would permit the oxidation of the skull material from melt refining to proceed at a fairly constant rate is being tested. In this system, the skull material is placed in a heated oxidation chamber which initially is filled with argon, and into which a constant flow of oxygen is admitted. A rise in pressure in the chamber signals a decrease in oxidation rate, whereupon a small amount of gas is discharged from the chamber to be replaced by oxygen. The resulting increase in oxygen concentration restores the desired burning rate.

A 500-gram charge of magnesium-zinc was vaporized in a melt refining furnace. Fiberfrax traps weighing about 2400 grams absorbed 95 percent of the vapors. The percentage of charge recovered in this experiment is similar to the percentage recovered in runs with Fiberfrax traps weighing about 1200 grams and charges of about 220 grams.

b. Development of Remote Methods of Fuel Fabrication - The original purpose of the program was to investigate remote control methods of fuel fabrication and to develop methods and equipment applicable to the EBR-II fuel cycle. The work was later extended to cover the preparation of installation drawings, job instructions and operational manuals for the EBR-II fuel recycle equipment. During the past month design work and procurement was completed on the following jobs:

- (a) Leak Detector Valve Cabinet.
- (b) Bond Tester Valve Cabinet.
- (c) Injection Casting Furnace Valve Cabinet.
- (d) Pin Processor Valve Cabinet.
- (e) Fuel Rod Assembly and Welding Valve Cabinet.

- (f) Vacuum Pressure and Cooling System for Injection Casting Furnaces.
- (g) Vacuum Pressure System Piping.
- (h) Pneumatic Argon and Control Piping in Subcell.
- (i) Electrical Work in Subcell.
- (j) Electrical Work in Basement.

Design work was completed, specifications prepared and procurement action taken on the fuel element assembly and welding machine, sodium meltdown and settling equipment and sodium bonding machines.

The vendors have now supplied about 98% of the components for the injection casting furnaces. The furnaces are being assembled for operational test before shipment. The five station pressure leak detector has been assembled and tested. Some difficulty has been experienced in achieving tightness with the "Adiprene" seals which replaced neoprene seals, formerly used. This is being corrected by a change in the hardness of the seal material and by refinishing of the seal surfaces. Major designs yet to be completed are the pulse-eddy-current bond and level inspection apparatus and the fuel element (subassembly) construction and welding machine. It is expected that drawings and specifications will be completed on both these machines in early March. Construction of the electrical controls and pushbutton stations for operation of the fuel reprocessing equipment is about 95% complete.

A furnace display and high frequency generator operating console was completed. This item and the furnace relay panel and push-button station were hooked up and placed in simulated operating condition. They performed satisfactorily. The pin process controls were similarly operated as were the welder controls. Modification to the welder controls was found necessary. Orders were placed for leak detection station instrumentation and for the components required to construct the fuel element assembly and welding machine controls. The data processor for the fuel pin process and inspection machine has been received.

c. Installation of Refabrication Equipment in the Fuel Cycle Facility - In the construction of the pneumatic and argon gas control valve cabinets, three cabinets have now been completed. Two more valve control cabinets have been started of a total of seven to be constructed. Minor gasketing problems have been encountered in valves and pressure regulators.

5. Fuel Development - Core I

a. Examination of Irradiated EBR-II Fuel Rods - Two additional NaK capsules each containing three U-5 w/o Fs alloy fuel pins jacketed

with 304 stainless steel and bonded with sodium have been opened after having been irradiated in the MTR. The two capsules, ANL-42-62 and 63, are part of a series which will provide irradiated EBR-II fuel rods for tests in the TREAT reactor. To date ten capsules in the series have been opened and the fuel rods examined. The results from the first eight capsules were reported in a previous Progress Report, ANL-6343 (March 1961), and in the Metallurgy Division Annual Report for 1961, ANL-6516.

Specimens in capsule ANL-42-62 had a maximum fuel temperature of 600°C, a maximum jacket surface temperature of 570°C, and a maximum heat flux of 609,000 Btu/hr-ft². The maximum burnup for the three specimens as determined by analyses of cobalt flux monitors was 1.7 a/o. The specimens in capsule ANL-42-63 had a maximum fuel temperature of 500°C, a maximum jacket surface temperature of 480°C, and a heat flux of 500,000 Btu/hr-ft². The maximum burnup of these specimens was 1.4 a/o.

The data from the six specimens from these two capsules substantiate and extend the results from the specimens previously reported.

Diameters and length measurements after irradiation were close to the reference values (0.174 in. D, 18 $\frac{3}{16}$ in. L). Preirradiation values were not available for direct comparison but measurable changes do not seem to have occurred within the precision of the measurements (length, ± 0.025 in., and diameter, ± 0.001 in.).

The bulk volumes of the jacketed specimens were calculated from immersion densities. The average volume increases for the specimens with 1.7 a/o and 1.4 a/o maximum burnup were 0.34% and 0.28%, respectively. These slight increases in volume are quite close to the precision of the measurements, i.e., ± 0.013 cc on the volume (7.4 cc).

All six rods were in excellent condition with no gross changes evident after irradiation. On the basis of the 30 rods examined thus far, the data indicate excellent stability of EBR-II fuel rods in the temperature range 360° to 600°C, with burnups to 1.7 a/o. Volume increases of less than 1% ΔV per a/o burnup can be expected.

A total of 345 enriched U-5 w/o F's alloy pins were to be made for tests in the TREAT reactor in enrichments of 5, 6, 7, 8, and 9%. Half of each batch of pins have been completed. The remainder have been assembled and bonded in EBR-II fuel jackets. Bond and sodium level inspection is under way.

6. Process Development

a. Melt Refining Process Technology - Additional information has been obtained on the nitridation rates of irradiated EBR-II-type fuel pins. The pins used in the experiments were 10 percent enriched uranium-5 percent fissium alloy irradiated to a burnup of about 1 total atom percent and cooled for 150 days. The nitriding atmosphere was 5 percent nitrogen-95 percent argon. Data obtained at 300°C were in agreement with results reported previously (see Progress Report, November 1961, ANL-6473, page 29). A single experiment at 650°C, the highest temperature investigated thus far, showed no temperature excursions or other effects that would indicate ignition.

Plans are in progress for a fifth melt refining experiment with highly irradiated fuel.

b. Skull Reclamation Process - Results of experiments in which uranium-fissium oxide and zirconium oxide were reduced concurrently by zinc-magnesium solutions in the presence of a molten halide flux indicate that uranium oxide is reduced more rapidly than zirconium oxide.

Decomposition of a uranium-zinc intermetallic compound at 650°C by the addition of magnesium resulted in a uranium precipitate in which the agglomeration of uranium was considerably less extensive than in decompositions carried out at 800°C.

Considerable effort is being devoted to devising and testing methods for the various phase separations encountered in the skull reclamation process. During the past month, work was concentrated on the separation of molten flux and metal phases. The performance of transfer tube units which accomplish separation of the two phases without requiring location of the interface was improved considerably by the incorporation of a metal phase disengaging chamber in the flux phase transfer line. Essentially complete separations of the phases were effected at practical transfer rates. Methods for the successive transfer of flux and metal phases were also investigated. These have the disadvantage of either requiring continuous location of the flux-metal interface and the ability to stop the transfer at a desired point or of providing an imperfect separation of the phases.

It has been confirmed that magnesium monouranate, $MgUO_4$, is the solid product resulting from the addition of uranium trioxide to molten lithium chloride-magnesium chloride mixtures (see Progress Report, January 1962, ANL-6509, page 24). A correlation appears to exist between the presence of the uranate and the reducibility of uranium oxides by zinc-magnesium alloy in the presence of halide flux

c. Blanket Processing - Equipment is being designed for concentrating the dilute plutonium product solution (plutonium dissolved in 50 percent magnesium-zinc) of the blanket process by evaporation of the magnesium and zinc. Equipment has been fabricated for the study of the removal of supernatant metal solutions from precipitated uranium metal, a phase separation which is required in both the blanket and skull reclamation processes.

d. Plutonium Recovery Process - Work on the removal of rare earth fission products from uranium-plutonium fuels by selective extraction into a molten halide flux is being continued (see Progress Report, January 1962, ANL-6509, page 25). Additional determinations of rare earth distribution coefficients between molten magnesium chloride and liquid magnesium-zinc alloys have confirmed earlier results for praseodymium, namely, that the distribution of praseodymium favors the flux phase in general, but that a minimum value for the distribution coefficient occurs in the region of 5 to 10 weight percent magnesium in zinc. In separate experiments, the behavior of cerium was found to be similar to that of praseodymium. The distribution coefficients were unaffected by a ten-fold change in rare earth concentration. Studies are in progress to determine the distribution coefficients of uranium and plutonium in the magnesium chloride-liquid magnesium-zinc system.

e. Reduction of Thorium Dioxide - Composition regions which are suitable for the reduction of thorium dioxide by magnesium-zinc solution have been defined for the calcium chloride-magnesium chloride-magnesium fluoride flux system. In general, the optimum region lies between about 8 and 14 atom percent fluoride ion concentration and above about 50 atom percent magnesium ion concentration in the flux.

f. Fused Salt Study - An absorption spectrum of uranyl chloride dissolved in lithium chloride-magnesium chloride at 650°C shows maxima at 675, 855, 1520, 1610, 1930, and 2230 millimicrons.

g. Materials and Equipment Evaluation - Tungsten continues to show excellent resistance to corrosion by the various molten halide flux systems encountered in the skull reclamation process. It is also showing excellent resistance to fluxes containing zinc chloride, mixtures which extensively corrode both tantalum and tantalum-10 percent tungsten alloy. A pressed-and-sintered tungsten crucible has been used, without visible damage, for nearly 300 hours of operations which simulate process operations.

Because it can be machined, a molybdenum-30 percent tungsten alloy is being considered for certain items of equipment, such as agitators and transfer tubes, for the skull reclamation and blanket processes. This alloy appears to have good corrosion resistance to molten zinc and flux

systems. An attack of 3 mils in 300 hours was noted on exposure of the alloy to pure zinc at 850°C; no attack by zinc-magnesium systems could be detected. An agitator shaft and blade made of this alloy has operated in process systems at 700° to 850°C for about 200 hours without evidence of attack.

D. Fast Reactor Test Facility (FARET)

The general engineering and physics parameters for an experimental facility to test characteristics of advanced fast reactors are being examined. These studies are directed toward a preliminary design of a test facility.

A study was made to determine the practicability of using bottom operated control rod drives. Although there are a number of advantages, particularly with respect to fuel loading and control rod management, there were sufficient disadvantages so that further consideration for the system has been terminated.

Various shielding components for the 150-cm (5-ft) extensions above the fuel assemblies were investigated. The materials of interest were (1) stainless steel; (2) alternate sections of 30-cm steel and 3% borated graphite; and (3) 90 cm zirconium hydride above 60 cm of steel. The results are shown in Table IX.

Table IX. FARET Fuel Element Shielding

Material in 150-cm Fuel Extensions	Flux at Top of Extensions, $n/(cm^2)(sec)$		Activation in 15-cm Cover after 1-yr Op- eration, mr/hr
	Fast	Thermal	
Stainless steel	3×10^{10}	7×10^7	500*
60 cm 3% borated graphite with 90 cm stainless steel in 3.30 cm layers	2×10^9	3×10^6	25*
90 cm zirconium hydride above 60 cm stainless steel	2×10^6	7×10^7	negligible

*Approximately one-eighth of saturated activity based on 500 ppm of cobalt in the stainless steel.

The total neutron flux streaming through sixty 0.16-cm ($\frac{1}{16}$ in.) wide annuli around the fuel extension was estimated. This flux, averaged over a diameter of 180 cm due to streaming, had a value of $4 \times 10^6 n/(cm^2)(sec)$. Therefore, additional shielding will not decrease the level of flux at the closure head due to the streaming.

III. GENERAL REACTOR TECHNOLOGY

A. Applied Reactor Physics

1. 3.0 Mev Van de Graaff

a. Experimental Studies of Neutron Scattering

(1) Scattering of Fast Neutrons from Ta¹⁸¹ and U²³⁸ - The elastic and inelastic scattering of neutrons from Ta¹⁸¹ and U²³⁸ is being studied throughout the incident neutron energy range 350 kev to 1.4 Mev. Using the Van de Graaff accelerator to generate neutrons, fast time-of-flight techniques (including an electromagnetic bunching system in the generator) are utilized to measure the velocity spectrum of scattered neutrons. The angular distribution of elastically scattered neutrons is determined at energy intervals of about 50 kev. The resulting differential elastic cross sections are expressed in the form

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_t(\text{el})}{4\pi} \left[1 + \sum_{e=1}^6 W_e P_e \right]$$

The excitation functions and angular distributions of inelastically scattered neutron groups are determined throughout the incident energy range. Inelastically scattered neutron groups are observed which correspond to excitation of the following nuclear levels: in U²³⁸ at 45 ± 3 , 150 ± 5 and 650 ± 50 kev; and in Ta¹⁸¹ at 150 ± 8 , 360 ± 10 and 510 ± 50 kev. These experimental results are being compared with those obtained by others, and with theoretical predictions based upon the optical model.

(2) Elastic Scattering of Fast Neutrons from Zn, Cu, Zr, and Nb - The angular distributions of elastically scattered neutrons from zinc, copper, zirconium, and niobium have been measured in the incident neutron energy range of about 350 kev to about 1.0 Mev, at intervals of about 100 kev. An electromagnetic bunching system, together with fast time-of-flight techniques, were used. This method enables one to obtain good energy resolution and to discriminate effectively against background and inelastic scattering. The experimental angular distributions have been fitted with a series of Legendre polynomials and the energy dependence of these coefficients, as well as the cross sections, have been compared with theoretical and other experimental results.

b. Theoretical Studies of Neutron Cross Sections - The NEARREX code has been completed. It is being used in conjunction with the ABACUS-2 and SCAT-IV optical model scattering codes to calculate consistent neutron cross sections for elastic scattering, polarization, inelastic scattering, capture, proton, and alpha particle emission and fission.

In particular, analyses of fast neutron interactions with yttrium, zirconium, and niobium were made. Measured elastic neutron scattering cross sections and polarizations in yttrium, zirconium, and niobium have been fitted with an optical model and the resulting transmission factors used to compute partial neutron reaction cross sections. The calculations employed the Hauser-Feshbach formula corrected for width fluctuation and interference effects and used the quadratic expression for transmission coefficients. Good agreement with available measurements of neutron inelastic cross sections was obtained and information on spin and parity assignments of levels was adduced where these are uncertain. Fitting of the niobium neutron capture cross section has been used to yield information on the energy and angular momentum dependences of the average level density and radiation width.

2. Instrumentation

a. Digital Method for Control Rod Calibration - A simple method has been developed for obtaining calibration curves for reactor control rods. It involves the digital recording of rod position and readings from two neutron counters while reactivity is decreased by driving the rod in from its critical position. A simple digital computer code then obtains reactivity as a function of rod position.

The recording system uses a Hewlett-Packard 560A 11-digit printer for readout. One of the digits records the contents of a 1-decade scaler which counts pulses corresponding to increments of rod motion. The remaining digits record the contents of two 5-decade scalars, which are connected to two neutron counters. All scalars are allowed to count for about $1\frac{1}{4}$ sec, after which the printer is energized. Counting stops during the 0.2-second printer cycle. After the cycle is complete, the scalars are reset and another interval of counting begins. A storage circuit is provided to hold any rod motion counts which come during the printer cycle. These are added into the scaler at the beginning of the next count.

This calibration technique was used on the JUGGERNAUT reactor (see Section II.A.5) where the rods are worth up to 1% Δk . Rod motion pulses were produced by a microswitch sensing the passage of teeth on a gear in the rod drive. Each neutron counter was placed in a hole in each of the two diametrically opposite thermal columns, in a position far removed from the reactor core, in order to minimize errors resulting from shifts in the fundamental mode. As a further precaution, the counts from each scaler were summed, and weighted so that each counter contributed equally. The use of pulse counters instead of the current chambers used with some analog methods simplifies precautions against mode shifts, since such counters are usable at much lower flux levels.

Rod calibrations obtained with the digital method agreed well with calibrations performed by other methods. The amount of reactivity which can be measured is limited by the fundamental mode shifts and by minimum usable count rates.

b. Rejection of Background Radiation Pulser in Hornyak Buttons - For counting fast neutrons in the presence of a gamma background, Hornyak button detectors have certain advantages over organic scintillators, especially a lower neutron energy threshold for a given degree of discrimination. The nature of pulses from alpha, beta, gamma, and neutron irradiation of Hornyak buttons has been examined under various conditions, including the use of light filters and with both fast and slow electronics. Tentative conclusions about the mechanism of their origin are that the pulses result chiefly from fluorescence induced by Compton recoils in the matrix and light transmitting media of the Hornyak button and phototube, with a secondary contribution from Cherenkov radiation. Such pulses are of short duration, being similar in shape to photomultiplier noise pulses, in contrast to the 70-nanosecond decay of the fastest component of ZnS(Ag) luminescence.

A relatively simple, passive anticoincidence network has been developed which allows fast cancellation of the gamma-induced short pulses while passing the zinc sulfide scintillation with adequate efficiency. Such a circuit is also useful where noise pulses, generated in the detection system, may be excluded without unnecessarily long integration. Rise time of the pulses emerging from the background suppression circuit is about seven nanoseconds. This network, in conjunction with a distributed amplifier, tunnel diode discriminator, and other fast electronic equipment is used to select fission neutrons in coincidence with fission events.

3. High Conversion Critical Experiment

Installation of the 1.18-cm triangular grid for Hi-C fuel (3 wt-% enriched UO_2) was completed. Criticality was reached with a small core having an atomic ratio, $\text{H}:\text{U}^{238}$, of 4.86:1. This core utilized about one-fourth of the available volume, and only three out of each seven grid openings in this small region are loaded. Insertion of fuel into the vacant positions will displace sufficient moderator to reduce reactivity. Reactivity may be regained by loading a larger fuel zone. This process will be followed cautiously until a limiting condition is encountered. Analytical studies indicate that the available fuel supply (5,574 elements) is insufficient to obtain criticality with every grid opening loaded. In this event fuel spikes or a more reactive second fuel zone may be added. The reactivity of the current loadings (550 to 600 fuel pins) agree fairly well with analytical predictions.

Analysis of measurements in the BORAX-V fueled cores continues. These were 5 wt-% enriched UO_2 elements in 1.27-cm square and 1.27-cm

triangular lattices. The atomic ratios, $H:U^{238}$, were 4.65 and 3.53 respectively. The central region measurements of ρ^{28} (subcadmium/epicadmium captures in U^{238}) were 5.49 and 4.98. Measurements of δ^{28} (total fission ratio U^{238}/U^{235}) were 0.065 ± 0.006 and 0.119 ± 0.010 . Comparisons with theoretical values¹ and other experimental values² gave good agreement with the 0.065 value but fell below the 0.119 result. The U^{238} capture distribution has been obtained for both cores.

4. Theoretical Physics

a. Doppler Coefficient Calculation - The determination of the magnitude of the Doppler coefficient in large plutonium-fueled fast reactors is of considerable interest because of safety considerations. The significant temperature-dependent cross section changes occur in the low energy region, below about 9 kev. Since leakage is small in this energy region, the Doppler effect depends essentially on two factors: (a) the relative capture in Pu^{239} and U^{238} of the neutrons slowing down below 9 kev; and (b) the fraction of total fission neutrons produced in the reactor slowing down below this energy. Because of the importance of elastic scattering and moderation by light elements in determining both of these factors, the ELMOE program is being used in conjunction with resonance integral calculations to solve the problems.

Among the factors being considered is the effect of sodium and structural material on the Doppler coefficient. A large part of the total reactor scattering cross section is provided by these materials. The presence of sodium is known to be important for the Doppler coefficient since 50 vol % increases the total low energy neutron source by a factor of 2. This means that careful consideration must be given to inhomogeneity effects caused by separation of fuel from sodium and structural materials in the design of experiments to measure the Doppler coefficient, either in critical experiments or in FARET. This problem may be acute in FARET where one would like to be able to use large fuel rods as a means of achieving high temperatures at low power densities. In the critical experiments there are also obvious advantages in not having the fuel as finely divided as in the reactor.

b. Fuel Cycle Studies - The original nine CYCLE production problems have now been rerun using the ELMOE code modified cross sections together with the conservative ν values for U^{235} . In all cases, critical masses and equilibrium cycle feed enrichments agreed closely with the original problems which used unmodified Yiftah, Okrent, Moldauer cross sections. These results indicate that the cross section refinements due to the ELMOE calculations are not very important for the systems now under investigation.

¹J. Bengston and R. L. Hellens, CEND-137, July, 1961.

²D. Klein, Z. Z. Krang, G. G. Smith, W. Baer, and J. De Juren, Nuc. Sci. and Eng. 3, 403-427, 1958.

Further production problems have been run using the ELMOE-derived cross sections. Cases studied have included the effect of U^{235} feed with no recycle on the 1000-liter metal system, 1750-liter oxide system, and the 437.5, 875, and 1750-liter carbide systems. Also, the middle size carbide system has been run for the case of higher Pu isotopic content feed, and for the case of Pu^{239} feed with single batch core operation.

The next series of problems to be run will examine the Th- U^{233} cycle using U^{233} feed. These problems will be of interest to the case considering the possible use of irradiated blanket elements for fuel elements in a Th- U^{233} thermal reactor core.

The associated COST code has been modified to take input from tape. The basic cost input data has also been slightly revised and expanded to consider the case of higher rental charges in addition to the original ten cost cases.

c. Feasibility of Pu^{239} - U^{235} Cores to Predict Pu^{239} Core Dimensions - Critical assemblies of plutonium-fueled dilute fast breeder reactors require large quantities of plutonium. Because of safety considerations and/or limited plutonium availability, it is desirable to determine the feasibility of employing composite Pu^{239} - U^{235} fueled criticals to predict properties of Pu^{239} fueled systems. For predicting the critical size, a cross-section parameter of the corresponding U^{235} -fueled critical might first be adjusted to force agreement of calculation with experiment. A cross-section parameter of Pu^{239} could subsequently be adjusted to force agreement of calculation with the observed size of the corresponding Pu^{239} - U^{235} fueled critical. The critical size of the Pu^{239} -fueled system might then be predicted with improved accuracy by use of the adjusted cross sections.

The case of homogeneous bare core mixtures having 5% fuel ($U^{235} + Pu^{239}$) by volume and 45% U^{238} , 16 $\frac{2}{3}$ % iron, and 19.3% aluminum as diluents has been studied. The decrease of the error in the calculation of the extrapolated bare core radius of the plutonium-fueled system depends upon the particular inherent cross sections which are in error and the particular cross section parameter (or parameters) which have been adjusted at the Pu^{239} - U^{235} composition. For the case of adjustment by ν^{Pu} at the composition ratio

$$\frac{Pu^{239}}{Pu^{239} + U^{235}} = 0.1$$

the error in radius is decreased by factors of about 8, 6, or 2 depending upon whether the inherent error is capture, inelastic transfer, or transport cross section.

d. Shield Design - EBR-II - The RE-122 multigroup diffusion code has been used with 20 groups to find flux distributions in a sodium pool behind 20 cm of uranium and also behind 20 cm of uranium preceded by 15 cm of steel. By determining the total boron capture distribution in the sodium, a comparison was made with the boron response measurements made in France* using a fission plate source and these same materials followed by 200 cm of sodium.

For the case where only uranium was in front of the sodium, the spatial distribution of the boron captures calculated from the 20-group fluxes was in good agreement with the measured response distribution. For the comparison with sodium absorptions, the calculated distribution was close to the experimental curve.

For the case where 15 cm of steel preceded the uranium, the calculated boron absorption rates decreased faster than the measured values at distances greater than 2 ft into the sodium.

B. Reactor Fuels Development

1. Corrosion Studies

a. Aluminum Powder Product Tubing - The original batch of Armour Research Foundation powder tubing (as-atomized A288 powder, compacted and extruded through a bridge die) has now been exposed to water at 290°C for 244 days, plus 30 days at 360°C. Also, two kinds of tubing made from as-atomized and from 70-hour milled powder have been tested for 279 days at 350°C. Sections of failed milled tubing have also been continued in test at 290°C for an exposure time to date of 230 days, plus 30 days at 360°C.

As of the end of February there has been no additional tubing failure. There is no appreciable end swelling of the material tested at 290°C. Since four batches of tubing of greater practical interest have been received and are being tested, no further corrosion evaluation will be made on the original material.

The eutectic bonded (Atomics International) plus "motor arc" welded tube end closures have been exposed for 292 days to water at 290°C. There have been no additional failures. Since these tube-closure combinations no longer represent material of current interest and are not believed to be the best that can be produced by current practice, tests have been terminated. It is encouraging that the relatively weak powder tubes which collapsed on the sharp edge of the end plug did not fail during corrosion testing.

*A. Kania, R. Pasquer, P. Pepin, M. Solanes, Paper SM-18/26, Seminar on the Physics of Fast and Intermediate Reactors, Vienna, August 1961.

Additional closure samples are being prepared.

Samples of finned powder product tubing, directly extruded by the Torrance Brass Company from as-atomized A288 billets, have been examined after 34 days exposure to 290°C water. Specimens are in good condition, with infrequent small blisters. These specimens have been added to a continuing test of powder product tubing at 290°C.

b. Zirconium Alloys for Superheated Steam - Analyses for hydrogen in corroded (in superheated steam) samples indicate two factors, in addition to composition, which seem to be important:

(1) Percentage of corrosion product hydrogen absorbed decreases sharply as the corrosion temperature is increased from 540° to 650°C.

(2) Heat treatment affects hydrogen absorption. It appears that annealing at 700°C increases hydrogen absorption.

The effect of temperature is not unexpected. At present we have no explanation for the effect of heat treatment. Evidence in support of these tentative conclusions is presented in Table X.

Table X. Effect of Corrosion Temperature and Heat Treatment on Hydrogen Pickup of Zirconium Alloys During Exposure to Superheated Steam (600 psi)

Alloy	Heat Treatment	Test Temp. (°C)	Time, days	Weight Gain mg/cm ²	% of Corrosion Product Hydrogen Absorbed
Zr-3 Ni-0.6 Fe	1	540	108	5.3	57
	1	650	88	9.4	14
Zr-1.1 Cu-1.1 Fe	1	540	108	6.9	44
	1	650	88	8.5	5.5
Zr-0.5 Cu-1.0 Fe	2	540	108	7.3	63
	3	540	108	4.8	25

Heat Treatments

- (1) Hot Rolled (900°C), Quenched (from 900°C), and Aged (700°C for 1 hr).
- (2) Hot Rolled (900°C), Cold Rolled, Quenched (from 900°C), and Aged (700°C for 1 hr).
- (3) Hot Rolled (900°C), Cold Rolled, Quenched (from 900°C), Cold Rolled.

2. Fabrication Development

a. Fabrication of Zirconium Alloy Tubing - Investigation is continuing of the unique lubricating properties of the red-grey coating mentioned

last month. This work indicates that copper-zirconium intermetallic compounds having a higher copper content than Zr_2Cu may be ideal drawing lubricants.

3. Ceramic Fuels

a. Preparation of Plutonium Carbide Fuels - The purpose of this program is to develop methods of producing plutonium monocarbide by direct reaction and to investigate means of producing usable specimens from plutonium carbide. Earlier results have shown that control of carbon content may be achieved by melting plutonium in contact with a carbon lined crucible and by careful control of temperature and reaction time. The process appears amenable to scaleup.

The equipment currently in use is a carbon element resistance furnace shown in Figure 7. The carbon heating element is formed by milling two equally spaced helical slots in a flanged carbon cylinder. The double helix so produced is attached by carbon screws to two conductive graphite support pieces. They, in turn, are connected to water-cooled mineral-insulated copper bus bars which pass through the furnace shell. The furnace shell is of the water-cooled stainless steel bell jar type. It is thermally insulated from the heating zone by five concentric reflectors - two tantalum, two molybdenum and one of heat resistant stainless steel. Various coil configurations may be used in this furnace up to approximately 6 in. inside diameter by 7 in. length. A maximum temperature of about $2100^{\circ}C$ was achieved. Power is controlled from a manually controlled step transformer. Temperatures are read by means of an optical pyrometer.

Figure 7

Carbon Coil Resistor Furnace

The brittle nature of the as-reacted carbide makes it very easy to pulverize into fine powder. These powders have been compacted into 0.156 in. I.D. specimen tubes using a vibrator powered by a 60-watt speaker driver and audio amplifier. Compaction of 60%-20 to 40 mesh, 20%-120 to 200 mesh and 20%-325 mesh powders have been compacted into 2 in. long, specimen tubes to a density of greater than 80% using a tamper rod. It has not yet been possible to produce samples which are free of activity in the top plug weld zone.

b. UC-PuC Bodies - Six separate runs of precipitated carbide were compared on the basis of composition and sinterability. The as-received carbide from each of the runs was ball milled dry for four hours. The milled material had an average particle size of 5μ with a maximum particle size of 15μ . The powders were each mixed with 3% naphthalene added as a solution with trichloroethylene. After evaporation of the solvent, the pulverized material was pressed at 15 tsi and the resulting specimens were vacuum sintered for two hours at 1750°C .

Table XI lists the compositions for each run and percent of theoretical density for specimens produced from each run. These data indicate that there is some correlation between free carbon content and densification. The larger the amount of free carbon the lower the density. It also appears that oxygen tends to interfere with densification under the conditions of sintering used. Runs 2 and 3 have similar C/U ratios but different oxygen contents and different densities. Run 5 has a higher C/U ratio than Run 2 but a considerably lower oxygen content with a higher density.

Table XI. Summary of Data on Precipitated UC Runs

Run No.	Method of Separating Free Carbon From Product	Product Yield %	Product Analysis				Sintered Density % Theoretical ^(c)
			C/U atom/atom	O w/o	Zn + Mg w/o	Fe + Ni + Cr w/o	
1(a)	Carbide allowed to settle, supernatant solution siphoned off.	68	1.27	0.90	0.5	0.01	80.0
2(a)	Carbide allowed to settle, supernatant solution siphoned off.	43	1.16	1.53	0.4	0.06	82.5
3(a)	Cropping ingot in reaction crucible	93	1.17	0.79	0.44	0.006	85.0
4(a)	Carbide slurried from reaction crucible.	47	1.26	1.17	1.3	0.03	81.5
5(a)	Melt partially frozen, supernatant siphoned off.	81	1.21	0.83	-	0.06	84.0
6(b)	Carbide allowed to settle, supernatant solution poured.	91	2.16	0.5	-	0.05	51.5

(a) Carbon source, activated charcoal (-200 mesh). Amount of carbon charged, C/U~1.5.

(b) Carbon source, powdered graphite (-200 mesh). Amount of carbon charged, C/U > 2.

(c) Pellets pressed 15 tsi, naphthalene binder, sintered 1750°C , 2 hours.

A new binder lubricant system was tried with the carbide and was found to be very satisfactory. It is a 2:1 mixture of acryloid and stearic acid dissolved in CCl_4 . Enough solution is added to make 3% by

weight solids to UC. The solvent is allowed to evaporate and the pulverized material is pressed in a die. The improvement in green density was from 6.9 gm/cc obtained with naphthalene to 7.9 gm/cc. Specimens pressed using this binder-lubricant have excellent green strengths and sinter to a smooth, good appearance. No difficulty was encountered in removing the binder-lubricant during the vacuum heat up.

4. Nondestructive Testing

a. Ultrasonic Techniques - Some time ago preliminary development work was undertaken on a high-power, short-duration pulse generator. This equipment, when completed, will produce a very short, well shaped pulse which will provide better resolution than the equipment now being used. It is hoped that a workable unit to generate, transmit, receive and detect pulses will be completed in the near future.

Studies are continuing on the wave guide theory of pulse propagation. No results are available yet from the exact theory; meanwhile, the problem is being examined with the help of an approximate model.

b. Neutron Techniques - Image sharpness studies are continuing and new test radiographs using gadolinium, scintillator, and rhodium direct exposure techniques are being evaluated for image sharpness qualities both on our own densitometer equipment and on a Leeds and Northrup recording densitometer. Further test radiographs obtained by transfer methods are planned now that CP-5 is once again in operation.

Contrast sensitivity capabilities using both direct exposure and transfer neutron radiographic methods are now under way, with the emphasis being to determine such values for heavy metals. Tests have been conducted thus far with uranium, lead, molybdenum and steel. With metal screen methods (either direct exposure or transfer) contrast sensitivities of the order of 2 to 4% have been consistently obtained and contrast sensitivity values of the order of 1% have been detectable in many cases.

Employing the faster scintillator techniques, contrast sensitivity values of the order of 6 to 10% were found. In spite of the poorer contrast capability of this method, its high speed makes it very attractive. For example, it has been possible to inspect up to 3 in. of normal uranium with an exposure time of less than 2 minutes or up to 6 in. of lead in about a 10 minute neutron exposure.

Further contrast sensitivity measurements on these and other metals are now in progress.

C. Reactor Materials Development

1. Radiation Damage in Metals

a. Neutron Damage Studies - Some experiments have been started to test the energy-dependent model for neutron damage to metals. The first irradiations include steel (SA-212B from a documented heat furnished by U. S. Steel) impact specimens and copper single crystals. Plans for the irradiations and predicted exposure rates were given in the Progress Report for December, 1961 (ANL-6485, p. 49).

Irradiations of steel in CP-5 began on February 12, 1962. One set is in the center of a fueled thimble element and the other in a thimble which is not surrounded by fuel. Before these irradiations began, jigs containing detector foils were exposed at these locations. Because the type of can used to contain the jigs did not permit it, no fissionable foils could be included in this preliminary exposure. Gold, nickel and sulfur foils were used and they are now ready for counting.

An identical set of steel impact specimens was loaded into the EBR-I. These are located in "wet" or NaK-cooled thimbles centered axially at the core midplane and occupying radial positions about 4.3 and 7.6 cm from the core centerline. Two single-crystal copper samples were also inserted in similar positions. The fragile copper specimens were sealed in stainless steel cans containing a small amount of NaK for a heat transfer bond. A nickel monitor wire was included with each specimen.

b. Magnetic and Dynamic Properties of SA-212B - The fundamental and third harmonic frequencies and the damping factors of the SA-212B modified magnet bars were measured over the temperature range between 80°F and 185°F for longitudinal (final rolling direction) and transverse specimens. Pre-irradiation measurements of the coercive force, at room temperature, of all bars have also been completed. The 60-cps hysteresis losses at several magnetic field strengths are being determined from photographs of the hysteresis loop. Similar post-irradiation examinations of the hysteresis loops will show irradiation effects on the following additional magnetic properties: permeability, remnance, and saturation induction.

c. Tuning Fork Irradiations - The tuning fork is a convenient device for the study of irradiation effects on the hardening mechanism and on the fatigue strength. Measurements of changes in fork frequency (which is related to modulus of elasticity) after predetermined exposures offers a convenient means of tracing the history of the radiation-induced hardening. Integrally weighted forks can be driven electrically, as cantilever beams, to establish damage effects on the fatigue strength. Forks of different frequencies can establish strain-rate effects on fatigue strength.

Studies are in progress to establish the most suitable geometric ratios between length, width, and thickness of tuning fork arms. The aim of these studies is to select a configuration free of harmonic and torsional frequencies.

D. Reactor Components Development

1. Development of Manipulators for Handling Radioactive Materials

a. Status of the Alpha-Gamma Metallurgy Research Cave (Building 212) - This unique shielded facility was designed so that research on highly toxic fuel materials could be carried out in a high purity atmosphere with a high degree of safety and efficiency. All research operations, as well as the repair of equipment, were to be carried out by the use of advanced types of remote handling equipment. These were to have included electrically-controlled master-slave manipulators supported from overhead, and radio-controlled slave-robots operating on the floor. In contrast to the conventional facility, personnel would not routinely enter the contaminated areas to perform such operations as set-up, dismantling, and repair of equipment, nor would the contaminated equipment be brought out for repair. In addition, if personnel are not required to enter, the lengthy and costly decontamination operations needed to reduce the penetrating radiation levels are eliminated.

A plan view of this facility for handling highly toxic materials, such as irradiated plutonium, is shown in Figure 8.

Figure 8. Plan View of the Alpha-Gamma Metallurgy Research Cave (Bldg. 212)

The main working area B, is connected to a number of service areas for the introduction of new samples, the preparation of waste materials to be transferred out, and facilities for the repair and storage of equipment. All the areas in which particulate dispersing material or equipment will be handled are essentially gas-tight. In order to minimize the number of transfers into and out of the enclosures, internal sample and equipment storage are provided for in the areas S and H. The main working area will be supplied with a recirculated purified gas atmosphere.

One of the original plans was to use sealed mechanical and electrically-controlled master-slave manipulators in the main working area. The electrically-controlled manipulators were to be supported in pairs by a bridge and crane system so that they could reach most of the working volume. This support system has been designed for ease of remote repair. A similar system will be used in area D, and a somewhat more complex interchange system will be used in areas E, F, C, and H. In addition to the overhead manipulators, slave-robots were to be used to help in the setting-up of equipment, repair of other equipment and other operations that would normally require personnel to enter in protective suits. The wheeled vehicle portion of the slave-robot would be radio-controlled and it would carry a pair of electrically-controlled master-slaves and television for viewing.

However, because of restricted funds the development of these advanced manipulator systems has been delayed. It will be necessary to initiate operations with remote handling equipment that can perform the immediate work, but at a slower rate and with frequent need for personnel to enter in protective suits. A General Mills Model 150 non-master-slave electric manipulator (40-lb capacity) will be substituted for the pair of electrically-controlled master-slaves mounted overhead. Also, for the initial operations a vehicle, powered and controlled through an electric cable and supporting a non-master-slave electric manipulator (about 100-lb capacity), will be used to transfer equipment and materials and to perform a limited number of adjustments and repair operations. It is estimated that the facility will be ready for limited operation with this initial equipment before the end of 1962.

The facility is designed so that new and improved remote handling equipment can be installed as it becomes available. If these new systems are incorporated, there will be the opportunity to test the concept of complete remote operation, and this operating experience will be invaluable in the development of future systems. The first step should be the use of the Mod. 4 Slave-Robot which will be completed in about 1.5 years. If up-dating continues to be carried out, the facility should improve in effectiveness for many years even though the research program may be continually changing.

E. Heat Engineering and Fluid Flow

1. Two-Phase Nozzle Tests

The experimental equipment to be used to investigate the effectiveness of a converging-diverging nozzle to convert the thermal energy in a two-phase water-steam mixture into kinetic energy is being assembled. Since the last report (ANL-6485, Progress Report for January, 1962), the heating section has been fabricated and the flow loop has been completed.

2. Boiling from a Liquid-Liquid Interface

An experimental investigation of nucleate boiling of saturated water and n-hexane from a mercury surface at atmospheric pressure has been completed. High speed motion pictures were also taken. A report on this work is being prepared.

The following conclusions were drawn from the study:

- (1) Nucleate boiling phenomena on a liquid surface is similar to boiling on a solid surface. The degree of superheat was, however, smaller in the latter case.
- (2) The position and slope of the nucleate boiling curve varied with the immediate history of boiling from a liquid-liquid interface.
- (3) On the basis of the data, it appears that boiling from a liquid surface is dependent on the chemical nature of the liquid-liquid interface and affects the degree of liquid superheat that can be attained at the surface.

A study will be made on the effects of foreign particles at the mercury surface on nucleation. The initial investigation will attempt to define the conditions under which a nucleation site becomes active in boiling.

3. Boiling Liquid Metal Studies

After careful cleaning procedures, the boiling alkali metal loop (see Progress Report, December, 1961, ANL-6485) components were welded into a single unit. Heaters, instrumentation, and insulation are being installed. High temperature tests of both the liquid level gage and the chromel-alumel thermocouples are being conducted for calibration and stability up to 1700°F.

4. Hydrodynamic Stability

Further tests were performed using the analog computer model to describe forced or natural circulation systems. The results have been

compared with data from the one heat transfer loop having two different diameter test section and riser combinations. The comparison of analog and experimental results on steam volume fraction and total flow rate for both steady-state and transient operation varied from good to excellent.

Under all conditions the analog model predicted the level of power at which the inception of instability could be expected (maximum error for the eight conditions tested to date was less than 10%) with good accuracy. Under those conditions where the analog model predicted the approximate proper slope of the steady-state flow to power curve, the analog prediction of the unstable power was excellent. For the six conditions where the analog predicted the proper slope, the prediction of the power at instability inception was within 1 kw of the measured value (within experimental measurement error).

The time-dependent behavior of a boiling loop is also being examined by determining the frequency response of steam volume fraction to flow and power oscillations.

The flow oscillator has been designed for the flow-to-void transfer function, and shop drawings are being prepared for fabrication of the unit. A turbine-type flowmeter will be used to measure inlet water velocity to the test section. The time response of the flowmeter signal converter was improved by a factor of four, to provide adequate response for the measurements.

The tests for the onset of hydrodynamic instability in a natural circulation loop are being continued using a 2.38 cm ID test section and riser having two restrictions in the single-phase region. On the completion of these tests the effect of orifice restrictions on the riser, and the effect of riser diameter, will be investigated.

5. Ruptures in High Pressure Steam-Water Containers

The maximum discharge of steam-water mixtures from breaks in vessel, pipes, and tubes are subjects of considerable concern in the evaluation of nuclear reactor accidents and containment problems. A program has therefore been initiated to study the flow of steam-water mixtures from tubes, pipes, and breaks in high-pressure vessels containing saturated or slightly subcooled liquid. The large-scale loop in Building 11 will be utilized for delivering highly pressurized water and steam to a 35-ft³ pressure vessel. Preliminary designs for installing the vessel, for the connections between the vessel and the large-scale loop, for the test sections, and for the apertures in the vessel wall have been completed. The variables expected to be studied are: discharge pressures over a range of 200 psia to 1500 psia, fluid qualities starting from 1%, and variation in the shape of the apertures. Methods for measuring flow rates,

pressure gradients, void fractions, and the metastability of fluid to be discharged have been outlined. From the experimental outline, it is believed that sufficient information can be obtained to develop models suitable for predicting flow rates from ruptures in vessels, tubes, and pipes under a variety of conditions.

6. Downflow Slip Ratio Study

A series of tests were performed in the large-scale loop test facility. The purpose of these tests is to determine pressure and velocity effect on downflow slip ratios. The range of variables covered in these tests were: mixture quality, X (0.003-0.14); superficial liquid velocity, V_s (0.8-4.0 fps); and system pressure (600, 1000, and 1500 psia).

The preliminary data indicated that there is little, if any, pressure effect on the downflow slip ratio when correlated as a function of the superficial velocity. The slip ratios approach zero as superficial velocity is reduced and approach unity as the velocity is increased. The study is to be completed shortly.

F. Chemical Separations

1. Fluidization and Fluoride Volatility Separations Processes

a. Fluoride Separations - The study of the removal of plutonium hexafluoride from air streams by combined vapor hydrolysis and filtration is being continued. Particle penetration data is being obtained for AEC-type filters. The information is being obtained in experiments in which small amounts, 25 mg or less, of plutonium hexafluoride are released in a chamber containing air of regulated moisture content. The air mixture is then passed through AEC filters, a water bubbler and Millipore filters all in series. Gamma and alpha counts of the filters and bubbler are used to determine the amount of penetration. It has been shown that when the air has the moisture content which is normally found under laboratory conditions (0.5 to 10 mg H_2O /liter) the AEC-type filter media is effective in removing the plutonium particulate matter formed when the plutonium hexafluoride is hydrolyzed in the vapor phase. As much as 99.99998 percent of the plutonium particulate matter incident on the surface of the first filter was removed from the air stream by two filters in series when the moisture content was 1.9 mg of H_2O per liter.

A sample of SF_4 was exposed to a gamma dose of 2×10^8 rads. Qualitative examination of the gaseous products by infrared spectroscopy indicated that S_2F_{10} was a major product and that a trace of SF_6 was formed.

It was demonstrated that mercury could react quantitatively with plutonium hexafluoride in a glass vessel at $25^\circ C$. Contact between the plutonium hexafluoride and the mercury was enhanced by shaking the

vessel. Following the reaction no unreacted plutonium hexafluoride was detected. Plutonium was recovered from the mercury by extraction with a solution containing 4 M nitric acid and 0.1 M aluminum nitrate. The exact form of the plutonium product has not yet been determined. The aqueous solution was analyzed for plutonium by alpha counting after extracting the solution with hexone.

Work has been started on a study of the chemical reactions of uranium chlorides. Of special interest are the reactions by which uranium chlorides are converted to uranium hexafluoride. The first reaction being studied is that of gaseous sulfur tetrafluoride and uranium hexachloride.

b. Direct Fluorination of Uranium Dioxide Fuel - Laboratory development work for the Direct Fluorination Process has been concentrated on experiments designed to elucidate the factors involved in the removal of plutonium by fluorination from mixtures of alumina, uranium oxide, plutonium oxide and fission product element oxides, which mixtures simulate the feed material in the fluorination step of the process. The presence of the fission product elements makes it more difficult to remove the plutonium from the alumina residues. The lowest residual concentration of plutonium was 0.04 percent, which was obtained by fluorination at 550°C for ten hours. It had been shown previously that pyrohydrolysis at 1000°C of fluorinated residues enhanced the removal of plutonium on further fluorination. It has been demonstrated that the same pyrohydrolysis treatment can be used followed by fluorination at 450°C for ten hours to decrease the plutonium concentration of a previously fluorinated alumina mixture from 0.29 percent to 0.02 percent. The pyrohydrolysis technique was most effective for residues containing 0.1 weight percent or more plutonium. At lower plutonium concentrations an additional fluorination at 450°C for ten hours was as effective as those fluorinations preceded by a pyrohydrolysis step.

Mixtures of uranium hexafluoride and plutonium hexafluoride can be separated by selective thermal decomposition. Plutonium would be deposited in the decomposition vessel as plutonium tetrafluoride and uranium hexafluoride would pass on through the vessel in the gas phase. In individual experiments at 200°, 250° and 300°C, gas mixtures having initial partial pressures of 90 mm UF_6 , 10 mm PuF_6 and 660 mm He were passed through vessels packed with nickel wool containing plutonium tetrafluoride. An average of 99.3 ± 0.3 percent of the plutonium was removed from the gas stream at each temperature and an average of 0.8 ± 0.3 weight percent uranium was deposited along with the plutonium. The gas volume was 9300 ml and the gas was circulated for four hours through the vessel at each temperature.

c. Fluorination of Uranium Dioxide Pellets - In the past, fluorination studies have been carried out with full sized uranium dioxide pellets ($\frac{1}{2}$ inch diameter by $\frac{1}{2}$ inch high cylinders) which are typical of the type that would be charged to a power reactor. During reactor operation, stresses developed in the pellets may cause extensive cracking, and it is likely that actual processing of spent fuel would involve pellet fragments as well as whole pellets. Accordingly a fluorination run was made in the three-inch reactor at 450°C with a 6-inch bed of cracked pellets. The pellet fragments measured about $\frac{1}{4}$ inch in diameter. Alundum grain was used as the fluidized material, nitrogen as the diluent gas, and the off-gas was not recycled. Because of poor heat transfer conditions, very little fluorine could be admitted without the temperature exceeding 500°C . The run was terminated after three hours of operation because of severe resistance to gas flow.

The third run in a series of deep-bed fluorinations (18 inches of $\frac{1}{2}$ inch diameter by $\frac{1}{2}$ inch long pellets) was carried to completion in 31.2 hours. Good temperature control at $490^{\circ} \pm 10^{\circ}\text{C}$ was demonstrated, and there was a complete absence of caking problems previously encountered in deep-bed fluorinations. Uranium hexafluoride production rates of 600 g/hr or more were maintained during the first two thirds of the run while the peak rate of 1530 g/hr was achieved midway during the run. Nine inches of alundum (static conditions) was used to enhance production rates during a major portion of the run. For a major portion of the run, fluorine input ranged from 25 to 30 percent of total gas flow while oxygen input ranged from 2.3 to 6.6 percent.

A uranium material balance in this run showed that the uranium hexafluoride condenser was about 96 percent efficient. This result also indicated that condenser efficiency is limited by refrigerant temperature (-40°C) rather than by fog formation.

d. Studies on Fluidized-Packed Beds in Mockup Systems - Fluidized-packed bed systems of process interest are being studied in support of the process development. The areas of interest include heat transfer, flow dynamics, and gas-solids mixing. Quantitative measurements of radial heat transfer have been made and it is planned to extend the measurements to axial heat transfer. Fluidized-bed expansion as a function of static alundum height and fluidizing velocity have been measured in a glass column; quantitative measurements of the elutriation rates of uranium oxide fines from fluidized-packed beds of pellets and alundum are also in progress with the glass mockup column. Preliminary experiments to measure gas mixing by tracer gas and thermal conductivity techniques are also in progress.

e. Plutonium Pilot Plant Facility - One of the two alpha enclosures for the plutonium pilot plant facility is on hand and the other is expected within a month. Most of the instruments have been ordered. Approximately

three-quarters of the equipment drawings have been completed and submitted for fabrication. A critical path (PERT) computation of the overall installation indicated that mechanical testing of all units can begin by the end of October 1962.

f. Studies on the Hydrochlorination and Fluorination Steps with Fixed-Bed Filters - A series of five runs has been completed in the $1\frac{1}{2}$ -inch diameter fluid-bed reactor on the hydrochlorination-fluorination reaction sequence for the processing of enriched uranium-zirconium alloy fuels. Fixed beds of alundum continue to give satisfactory results as high-temperature filters for retention of uranium particulate entrained by the zirconium tetrachloride off-gas stream produced during hydrochlorination. The first of the current series of runs used the combined up-flow and down-flow filter bed system while subsequent runs used only a down-flow filter bed. Uranium losses during hydrochlorination were less than 0.2 percent of the charge which averaged 16.7 g as 4.83 weight percent uranium-Zircaloy alloy chips ($\sim\frac{1}{4}$ inch on a side). Re-use of both the reaction bed and filter bed was made for the entire series of five runs and uranium content of the beds after fluorination indicated that the cumulative residual uranium can be reduced to nearly one percent with such re-use.

g. Fluid-Bed Hydrolysis of Zirconium Tetrachloride - Modifications to the six-inch diameter fluid-bed system being used for studying the zirconium tetrachloride hydrolysis with steam were made to increase the capacity to the flowsheet specifications, i.e., two kilograms of tetrachloride per hour. The use of a Vibra-Screw feeder and air conveyor system achieved the desired rate of tetrachloride solids feed. Excessive pressure drops across the sintered metal filters located directly above the bed indicate that residence time of the reactants in the bed zone may be insufficient and hydrolysis may be occurring on the filters.

2. Chemical-Metallurgical Process Studies

a. Liquid Metal Solvent Studies - Additional measurements have been made on the solubility of uranium in liquid magnesium. The following values (in weight percent uranium) were obtained: 0.36 at 846°C, 0.53 at 902°C, and 0.64 at 948°C. These values are in substantial agreement with the values reported by Chiotti and Shoemaker.¹

The solubility of zirconium in about 50 weight percent zinc-magnesium solution has been measured. The following tentative values (in weight percent zirconium) were obtained: 0.02 at 422°C and 0.49 at 709°C.

¹P. Chiotti and H. E. Shoemaker, Ind. Eng. Chem. 50, 137 (1958).

Measurements of the solubility of praseodymium in liquid zinc have been extended over a wider temperature range by the use of a radio-chemical method. The solubility may be represented over the temperature range 454° to 713°C by the linear equation

$$\log (\text{atom percent praseodymium}) = 6.447 - 6764 T^{-1}$$

The distribution of zirconium-95 between lead and zinc has been measured. The values of the distribution coefficient were found to be 531 at 701°C, 350 at 732°C, and 106 at 757°C. It should be noted that the low level of activity of zirconium-95 in the analytical samples introduced a large uncertainty in the results. Therefore, the values obtained should only be considered as approximate.

Results obtained in the further study of the lanthanum-zinc system by means of the continuously recording effusion balance agree substantially with the preliminary results reported earlier (see ANL-6409, Progress Report for August, 1961, page 57). However, slight corrections in the stoichiometry of some of the phases were considered necessary. The present data indicate the existence of the following phases: LaZn_{13} , $\text{La}_2\text{Zn}_{17}$, LaZn_2 , LaZn_{11-12} , $\text{LaZn}_{7.3-8.0}$, $\text{LaZn}_{3.8-6.4}$ and LaZn . The first four of these phases have been confirmed by X-ray diffraction analyses.

Preliminary results obtained in a study of the neodymium-zinc system by the effusion method have revealed the existence of the following phases: $\text{NdZn}_{8.5-11.5}$, Nd_2Zn_9 , and probably NdZn_3 .

b. Calorimetry - Exploratory experiments have been carried out in preparation for the calorimetric study of tantalum and niobium in fluorine.

Exploratory, noncalorimetric experiments were also carried out with sulfur, carbon, and zirconium diboride. Both sulfur and zirconium diboride were found to ignite spontaneously in fluorine. In the case of zirconium diboride, the combustion temperature was high enough to initiate the combustion of the nickel dish which supported the sample. Powdered graphite did not react spontaneously with fluorine, but could be ignited readily by means of an electrical fuse wire.

3. General Chemistry and Chemical Engineering

a. Conversion of Uranium Hexafluoride to Uranium Dioxide

(1) Simultaneous Reaction Studies - A brief study of the simultaneous reaction scheme (steam and hydrogen at 650°C) for converting uranium hexafluoride to uranium dioxide is being made in attempts to produce dense particles suitable for reactor fuel (dispersion, or compacted-tube elements). Run conditions were a uranium hexafluoride feed rate of

10 g/min, steam-hydrogen ratio of 2, six-kilogram starting bed, 0.75 ft/sec superficial gas velocity and a bed temperature of 650°C. Only a slight increase in particle density, 6.4 to 6.6 g/cc was noted in initial trials; the residual fluoride content of the product dioxide was encouraging, ranging from 150 to 1000 ppm.

(2) Two-Step Process - Exploratory studies on the higher temperature hydrolysis step for reacting uranium hexafluoride (500°C as opposed to 200°C) showed that the reactor coating problem could be essentially completely eliminated. Chemical analysis of the varied-colored products indicate that some uranium tetrafluoride may be present. Color changes and bulk density increases (from 2.1 to 3.1 over a 6-hour period) were associated with greater conversions to U_3O_8 . Elimination of uranyl fluoride as the primary reaction product apparently reduces the amount of the coating. A three-hour batch test showed that uranyl fluoride is converted readily to U_3O_8 at 500°C with steam.

b. Fluid-Bed Calcination Studies in Small-Diameter Columns - A new type of two-fluid feed spray nozzle is being examined. The previous nozzle was an extended tip nozzle with a protective air cap. The new nozzle is a submerged tip liquid nozzle. It is similar to the previous nozzle except that the internal air orifice has now been removed (see Figure 9). Satisfactory runs ranging in duration from 3.7 to 14.0 hours were made at substantially increased flow rates, 20 ml/min compared to previous rates of approximately 10 ml/min. These feed rates give a correspondingly large decrease in the air-to-liquid ratio of atomizing gas to liquid feed (from 2000 to 600). This is considered important, since a secondary objective of this work is the achievement of overall reduction in off-gas volumes from calciner-type operations. Current experiments are directed toward establishing the reliability of the process and achieving remote operation.

Figure 9. Two-Fluid Nozzles

c. Preparation of Uranium Carbide - Investigation of various methods for the preparation of uranium carbide was continued. In one experiment, excess carbon was added to uranium dissolved in zinc-magnesium

solution, and the mixture was held at 800°C for a total of 25 hours. Uranium carbide was the only product found. The addition of several hydrocarbon gases to uranium dissolved in cadmium-one weight percent magnesium solution appeared to form some uranium carbide, but the reaction rate under the specific conditions was too low for practical use.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Direct Conversion Studies

1. Plasma Oscillations in a Thermionic Potassium Cell

a. Experimental Setup - The relationship of the r-f oscillations and the d-c output of the potassium plasma cell was investigated experimentally. Figure 10 shows a schematic of the experimental arrangement. As seen in Figure 10 one r-f choke is placed between the collector and applied voltage source circuitry and the other choke between the collector and the x-y recorder. A $2\ \mu\text{F}$ capacitance C connects the collector to a scope and vacuum tube voltmeter. The choke capacitance network separates the r-f from the d-c output. The r-f voltage can be measured across an open circuit or load, or the r-f can be grounded.

Figure 10. Voltage-Current-Measurement

The voltage-current characteristics were traced with an x-y recorder. The voltage applied to the collector and guard ring was obtained from a transistorized inverter circuit as reported in ANL-6473 (Progress Report for November, 1961, p. 63).

b. Experimental Procedure and Results - The following parameters could be varied in the experiments: (1) emitter temperature; (2) potassium vapor pressure and atomic density; and (3) the connections to the r-f circuitry. Figure 11 shows the curves for the d-c voltage-current characteristics as functions of temperature, applied voltage, potassium pressure, with r-f open (0), and r-f short-circuited connections (S). The scale to be

used is marked at the right of each curve and also the multiplication factor for the figures of the scale. The frequencies, in kc, are recorded for applied d-c voltages: +3, +1, 0, -1, and -2 volts. Table XII shows the potassium pressure, the vapor density at the trap temperature in atoms/cc, the emitter temperature, and the measured RMS open-circuit voltages. Column 10 of Table XII shows the power maxima for d-c (PMDC) obtained from the product of the voltage and the current at the left knee of the curve. The power maxima for r-f (PMRF) were obtained by inserting various inductance-free resistances (r-f load in Figure 10) between the r-f input and ground and varying the d-c applied voltages from zero (short circuit d-c) to -2 volts. The PMRF usually occur when the d-c voltage is close to zero and it is seen that the frequency decreases with a decrease in the applied d-c voltage. In Table XII it is seen that for the lowest temperature and current, curve "a," no r-f output is generated. For curve "b," "c," and "d," the open and short-circuit characteristics vary considerably. As shown in Figure 11, curve "d," the d-c current at open r-f is only 55% of the d-c current with shorted r-f. An increase in temperature or in d-c current decreases the frequency with a constant potassium pressure.

Figure 11. Potassium Cell

Table XII. Schedule of Experimental Results Shown in Figure 11.

1	2	3	4	5	6	7	8	9	10	11	12
	P(K) mm Hg	No(K) AT/cc	Em-T °K	AT + 3 Vc RMS-V	AT + 1 Vc RMS-V	AT-0-Vc RMS-V	AT-1 Vc RMS-V	AT-2 Vc RMS-V	PM DC	PM RF	OP-C DC-V
a	2.5×10^{-4}	6×10^{12}	1950	—	—	—	—	—	2 mw	—	3.2
b	2×10^{-4}	4.8×10^{12}	2030	0.26	0.37	0.46	0.48	0.37	7.4 mw	0.3 mw	3.4
c	2×10^{-4}	4×10^{12}	2170	1.8	2.7	2.3	1.75	0.9	36 mw	3.5 mw	3.6
d	1.9×10^{-4}	3.9×10^{12}	2300	6.0	4.9	3.2	2.3	1.0	143 mw	36 mw	3.7

Figure 12 and Table XIII show the curves and data for two higher emitter temperatures.

Figure 12. Potassium Cell

Table XIII. Schedule of Experimental Results Shown in Figure 12

1	2	3	4	5	6	7
	P(K) mm Hg	No(K) AT/cc	EM-T °K	PM DC	PM RF	OP-C DC-V
a	3.8×10^{-4}	4.5×10^{12}	2380	288 mw	65 mw	3.8
b	3.8×10^{-4}	4.5×10^{12}	2520	1.1 W	50 mw	3.7

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Fuel-Coolant Chemical Reactions

The series of condenser discharge runs with aluminum wires in water at 200°C was concluded. An unusually slow pressure rise (two seconds) was confirmed. This was more indicative of burning propagation from a hot spot than it was of the reaction of small molten particles. Further studies are underway in which the nature of the reaction will be studied photographically.

Studies of metal-water reactions by the pressure-pulse method are continuing. Additional runs with aluminum-5 percent uranium alloy have been completed.

Six experiments have thus far been performed in TREAT on shelf type, graphite fuel specimens. A summary of the conditions and results is shown in Table XIV.

Table XIV. Treat Runs on Graphite Fuel Specimens

<u>Conditions</u>				
Graphite fuel pin at 25°C at start of transient Fuel pin in a 20 psia helium environment Reactor periods of about 100 milliseconds				
Energy Input to Fuel Specimen (Arbitrary Units)	Peak Temp of Fuel Pin during Transient (°C)	Appearance of Fuel Pin after Transient	Mean Particle Diameter, d _{sv} (mils)	Microcuries of Xe ¹³³ Released from Fuel ^a
1.2	850	intact, no change	-	6
2.5	1700 ^b	some cracking at top, but retained basic cylindrical shape	-	34
several transients ^c	-	destruction into fragments and particles	62	4700
3.9	2510 ^b	destruction into fragments and particles	32	1400
6.5	2900 ^b	destruction into fragments and particles	23	2100
8.9	3350 ^b	destruction into fragments and particles	22	2300

^aMeasured from gas sample of autoclave and corrected back to time of irradiation.

^bObtained from calibration run of temperature versus energy input.

^cFrom the plot of observed temperature versus reactor energy there is a slight "knee" in the curve at about 2.0 to 2.5 energy units thus suggesting break up of the fuel in that range.

These results indicate that the maximum allowable fuel temperature, corresponding to the threshold of destruction, is about 1700°C. Transients which are energetic enough to cause peak graphite temperatures greater than this value cause fragmentation of the fuel. Very little xenon-135 is released in nondestructive transients relative to the destructive ones.

An energetic TREAT transient of 750 megawatt-seconds on a 39-millisecond period gave 19.0 percent reaction of molten aluminum with water.

2. Metal Oxidation and Ignition Studies

The calculations of burning temperatures of metals in air is continuing. Previous calculations (December 1961 and January 1962 monthly reports, ANL-6485 and ANL-6509) for uranium and zirconium burning in air assumed a natural convection process. Calculations, extended to include forced convection, indicate that a linear air velocity of about 10 cm/sec or more is required before a significant increase in burning temperature results from forced air circulation for air velocities as high as 3000 cm/sec.

Recent high temperature studies of the isothermal oxidation of uranium have been used to compute burning curve ignition temperatures. The method is a stepwise calculation of the time-temperature history of uranium specimens during programmed heating. Graphical rate data and a simple heat transfer model are used in the calculations. Calculated and experimental results for 9.5-mm uranium cubes are in excellent agreement.

Evidence has been obtained for the existence of oxides higher than $\text{UO}_{2.0}$ on the outer surface of oxidized uranium. Further work is planned to establish more precisely the time at which formation of this material begins.

A halogenated hydrocarbon is being sought for addition to Freon 152 which will result in a nonflammable mixture that retains the effectiveness of Freon 152 for suppressing the burning rate of uranium and zirconium. Both CF_3Cl and CF_2Cl_2 are currently under study.

Vacuum-shielded ignition studies on spherical zirconium metal powder are proceeding. The effects of exposure to air and effects of varied sample size are being studied.

Recent results of the ignition and oxidation studies of plutonium show that the oxidation follows a linear rate law in the beta and lower gamma phase temperature range. The parabolic rate law is followed in the alpha, upper gamma and delta range.

B. Fast Reactor Safety Studies

1. Experimental Meltdown Program

In-pile meltdown experiments are being performed in the TREAT reactor in order to study fast reactor fuel failure, the resulting movement of molten fuel, and the mechanisms producing failure and meltdown product movement.

a. EBR-II Element Experiments in the Absence of Sodium - Experiment Series XXIX was prepared and shipped to TREAT. This is the first series of samples designed to study failure and meltdown product movement resulting from exposure of elements arranged in clusters. Each capsule was loaded with seven natural enrichment EBR-II Mark-I elements on an EBR-II pitch with one central element surrounded by a ring of six. Tantalum neutron absorbers were contained in the capsules to produce axial power profiles typical of the EBR-II core. A calibration capsule and two of the four meltdown experiment capsules were run at TREAT during the report period. Exposure of the remaining two capsules was delayed pending analysis of the transient temperature records from the first two capsules.

Relative sample activation of the clustered elements in the calibration capsule was obtained by gamma counting. Both the central element and an element from the surrounding ring displayed an activation profile of the desired "chopped cosine" shape, and maximum-to-minimum ratio. Readings from the outer element were about 4% higher than those from the central element, in agreement with predictions. Samples have been submitted for radiochemical determination of total sample fissions.

Temperature records from the two meltdown capsules analyzed showed that central element temperatures tended to "coast" upward beyond the outer sample temperatures, after the initial sharp temperature rises. Time scales for the differential rise were consistent with those expected for the differences in heat transfer during the tail of the reactor power pulse. The central element is surrounded by a ring of hot elements, while the outer elements are surrounded by the graphite meltdown crucible which remains near room temperature. The temperature records indicate that temperature rises during the early "adiabatic" sample energy input are close to those predicted on the basis of calculated sample flux depressions. (A direct comparison of predicted temperatures and maximum central element temperatures would imply the calculated power calibrations were about 35% low.) Accordingly, the TREAT specifications for the two remaining capsules have been set for shorter power pulses to produce a nearly adiabatic energy input up to the time of failure.

b. EBR-II Elements in Stagnant Sodium - The stagnant sodium capsule experiments of Series XXVII were examined during this period. This series consisted of four EBR-II fuel elements, each individually loaded into a stagnant sodium capsule and exposed to transients of differing severity and with the flux axially tailored along the element. The test conditions for these elements are listed in Table XV.

Table XV. Axially Tailored Power Stagnant Sodium Experiments

Sample No.	Initial Reactor Period (ms)	Integrated Reactor Power (Mws)	Maximum Fuel* Temperature (°C)	Sample Condition
1	109	152	680	Slight warp, little eutectic.
2	110	197	740	Eutectic midsection, approximately 20 cm.
3	89	251	810	Failed, section gone 12 cm from bottom, many penetrations along length.
4	92	243	390	Failure similar to No. 3 but greater in extent.

*Thermocouple embedded in end of fuel pin, in region of low sample power.

Because of the increased neutron flux depression of the sample produced by the tantalum neutron absorbers used to shape the power, samples 3 and 4 were run at higher initial k_{ex} (shorter initial periods) in order to obtain failure. It was observed that these exposure conditions produce a failure pattern which is less localized than that found when similar samples were run in dry capsules. (See Progress Report for January, 1962, ANL-6509.)

c. Dry Opaque Capsule for Remote Assembly - The equipment for the remote fuel encapsulation of irradiated samples in dry opaque capsules has been received. The coffin was tested by drawing a specimen of spent CP-5 fuel into the cavity. External radiation surveys showed that the unit is entirely satisfactory for use.

The thermocouple spot welder was tested and demonstrated the capability of securely fastening the thermocouples to the cladding.

Equipment handling and mockup studies are underway and it is expected that actual capsule assembly will begin in the near future when the capsule components are received.

d. Dry Transparent Capsule - Remote Loading - Design and testing have been finished on modifications to the dry transparent capsule to permit remote loading with irradiated elements. Handling accessories for the remote loading are being constructed. Work has begun on design specifications of modifications to the steel slot liner which contains the transparent capsules in TREAT, and on a shipping coffin for the capsules.

e. Small (Package) Sodium Loop - The handling coffin design proposal for the small package sodium loop has been reviewed and found satisfactory.

A total of three package loops have been almost entirely assembled. The one item of difficulty has been the pressure transducer, which was found to be unduly sensitive to temperature gradients and subject to damage. As a consequence, considerable delay was experienced in the installation of two of these units on the loop. However, this has now been accomplished and the loops are now being prepared for the first out-of-pile tests. The instrumentation for operating the control has been designed. The purchase of instruments has been initiated and delivery is scheduled for March 26, 1962.

f. Large Sodium Loop - The drawings required for the TREAT building modifications have all been completed. The cold trap, plugging indicator, and gas transfer system designs are approximately 50% completed. In addition, wiring and heater installation requirements are under study. A grinder to shape the pipe ends for welding has been designed. Expansion joint and test section of expansion joint types to be used are under consideration.

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