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Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

September 1964

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

September 1964

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FOREWORD

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

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

The last six reports issued
in this series are:

March 1964	ANL-6880
April 1964	ANL-6885
May 1964	ANL-6904
June 1964	ANL-6912
July 1964	ANL-6923
August 1964	ANL-6936

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I. BOILING-WATER REACTORS

A. BORAX-V

1. Operations

The BORAX-V reactor and facility have been shut down and the experimental program terminated. All steam, water, and pneumatic systems, tanks, equipment, and instrumentation have been drained and dried. The power to all operating and experimental instrument and electrical systems, as well as the building heating systems, has been secured. All boiling fuel assemblies are stored in the dry boiling fuel-storage pit. The highest central UO_2 temperature from decay heat measured in a dry boiling fuel thermocouple rod was 235°F . Thirteen peripheral superheater fuel assemblies are stored in their normal locations in the dry reactor vessel, and the remainder of the peripheral and central superheater fuel is stored in the dry superheater fuel-storage tank. The highest measured superheater fuel temperature in dry storage was 230°F . Additional concrete shielding slabs have been placed over both fuel storage pits.

All reusable special purpose reactor components, experimental equipment, and spare parts have been stored at the reactor site. All reusable general purpose experimental equipment, instruments, spare parts, and tools have been transferred to other ANL projects.

Four inboard 4-plate fuel elements, two each from an inlet and an outlet peripheral superheater fuel assembly irradiated in core PSH-1, and a boiling fuel thermocouple rod irradiated in cores CSH-1 and PSH-1 have been shipped to Argonne, Illinois, for detailed hot cell examination. Underwater examination of the ends of the superheater fuel elements revealed no observable fuel-plate distortion. The average burnup of each of the four superheater fuel elements ranges from 0.45 to 0.61 a/o of the U^{235} in the stainless steel- UO_2 cermet plates. The average burnup of the boiling fuel thermocouple rod is 0.074 a/o of the total uranium in the UO_2 fuel.

To disassemble the radioactive (about 100,000 r/hr) superheater fuel assemblies prior to placement in a shipping cask, a special remote-control underwater hacksaw was designed and fabricated. The hacksaw is powered by a pneumatic motor; and long-handled socket wrenches are used to actuate saw feed, saw blade changing, upper and lower clamp jaws, and slicing fixture elevation. Three cutting operations were performed on each superheater assembly. The bottom nozzle and top riser were first removed with the fuel assembly in a vertical position. Then the remaining 5-element fuel subassembly was laid horizontally in the slicing fixture and the individual fuel elements separated by slicing through the flow vanes connecting the insulating tubes.

A 1-in.-square, prepared, defective superheater fuel sample was removed from the coolant channel of an outlet fuel assembly and shipped to Argonne, Illinois, for hot cell examination. Stainless steel and aluminum alloy corrosion samples which had been mounted on the outside of the structures of all three BORAX-V cores were also shipped to Argonne, Illinois, for examination.

2. Boiling Fuel-rod Central Temperatures

Temperature data from operation of boiling thermocouple fuel rod No. 17, Core Position 45, with core PSH-1 are shown in Figure 1. This rod containing a W-W/26 w/o Re thermocouple in the radial and axial center of the UO_2 pellet fuel, had been subjected to thermal transient tests when new and then irradiated during BORAX-V operation with cores CSH-1 and PSH-1 for an average burnup of 0.074 a/o of the total uranium in the 4.95 w/o enriched UO_2 fuel. (Only 55 MWhr of total integrated power accrued during operation with core CSH-1.) The indicated fuel temperatures show the same general pattern as that reported for other 4.95% enriched rods (see Progress Report for August 1964, ANL-6936, pp. 10-13; Progress Report for July 1964, ANL-6923, pp. 7-8): increased burnup leads to increased fuel temperatures up to a point where temperatures first reach the region of about 2200°F. Beyond this point, the pattern becomes erratic and unpredictable. To verify that the indicated temperatures were not in error, transient and static post-irradiation calibration tests were performed.

The first tests measured the overall thermal time constant of the irradiated fuel rod and thermocouple, and duplicated tests previously made on some of the fuel rods when in a new condition (see Progress Report for September 1963, ANL-6784, pp. 3-4). The tests were made by inserting two thermocouple fuel rods, from room temperature conditions, into NaK at 500°F and recording the output from the fuel thermocouples. Total fuel rod immersion time was measured by using a continuously indicating resistance level-detector, and was found to require 0.7 to 1.0 sec from the time of initial NaK contact. The 63.2% thermal time constants, determined in this manner, were found to average 13.1 sec for the irradiated rod VA-2 and 7.8 sec for rods VA-1 and VA-2 when new.

In an attempt to simulate the transient test findings, a thermal analog model was set up and tested by use of an analog computer. Typical values of thermal resistances and heat capacities were tried in various combinations to get approximations of the measured thermocouple-fuel-rod time response. Manipulation of the circuit elements was used to study the relative effects of changes in resistance of the cladding and "film," thermal resistance of fuel and the thermocouple time constant. Since the steady-state, in-reactor fuel temperature was not governed by heat capacity and thermal resistance values associated with the fuel rod thermocouple, the primary effects studied on the model were those of variations in the fuel and cladding parameters.

Figure 1. Boiling Fuel Rod Central Temperature vs. Reactor Thermal Power and Burnup, for Core PSH-1 of BORAX-V

The analog circuit representing the thermocouple fuel rod is shown in Figure 2. Some results of the model study are compared in Figure 3 with actual tests on the fuel rods. The relatively large initial delay in the fuel thermocouple response (curves 1 and 2) is believed to be due primarily to a "film" resistance resulting from a lack of wetting of the stainless steel fuel clad by the NaK. Time zero is taken as the point of initial contact of the fuel rod bottom with the NaK. Other studies made with the model are summarized in Figure 4 and Table 1.

R_1 = Thermal Resistance of Clad

R_2 = Total Thermal Resistance of Fuel and Fuel-to-clad Gap

R_3 = Effective Combined Thermal Resistance of Fuel-to-thermocouple Gap and Thermocouple

C_1 = Total Heat Capacity of Clad

C_2 = Total Heat Capacity of Fuel

C_3 = Total Heat Capacity of Thermocouple

E = Initial Temperature Differential between Thermocouple and Heat Source. (Model Assumes a Step Increase in Temperature)

V_{out} = Temperature of Thermocouple

Figure 2. Analog Thermal Model for Boiling Thermocouple Fuel Rod of BORAX-V

Figure 3. Computed and Measured Thermal Time Response of Boiling Thermocouple Fuel Rods of BORAX-V

Figure 4. Computed Thermal Time Response of Boiling Thermocouple Fuel Rods of BORAX-V

Table I. Constants Used in Analog Thermal Model Studies

Run No.	R_1	C_1	R_2	C_2	R_3	C_3	$\frac{R_2 C_2}{R_1 C_1}$	$\frac{R_2}{R_1}$	$\frac{C_2}{C_1}$	τ , sec
A	5	0.2	10	1	10	0.1	10	1	5	12.8
B	5	0.2	5	1	10	0.1	5	1	5	7.6
C	2	0.2	5	1	10	0.1	12.5	2.5	5	6.9
D	10	0.2	10	1	5	0.2	5	1	5	13.5
E	0.1	0.2	10	1	5	0.2	500	100	5	12.1

NOTES:

1. Resistances in meg, capacitors in μf .
2. Time constant τ is time required to make 63.2% of total change in temperature from time zero.
3. Based on original fuel design, the constants were calculated to be related by:

$$\frac{R_2 C_2}{R_1 C_1} \equiv \frac{RC \text{ (Fuel)}}{RC \text{ (Clad)}} = 1050. \quad \frac{R_2}{R_1} \equiv \frac{R \text{ (Fuel)}}{R \text{ (Clad)}} = 244.$$

$$\frac{C_2}{C_1} \equiv \frac{C \text{ (Fuel)}}{C \text{ (Clad)}} = 4.3$$

The effective time constant of the thermocouple is of the order of 0.5 to 1 sec (in the pre-irradiated condition).

Transient test with fuel rod VA-2 confirmed the in-reactor, static temperature indications of an approximate 50% reduction in the thermal conductance of the fuel due to fuel burnup and operation at elevated temperature. The analog studies were limited, and exact matching of the model and physical case was not obtained. Very similar time-response curves and 63.2% time-constant values were obtained with the model. Furthermore, changes in the thermal resistance of the fuel are much more important to the overall time constant than changes in fuel cladding or film resistances because of the small value of the ratio of cladding heat capacity to fuel heat capacity. (The overall time constant is directly related to the "RC" time constant values of the sections of the fuel rod.)

The accuracy of the thermocouple in fuel rod VA-2 has been confirmed by pre-operational, in-reactor, zero-power calibrations and by a post-irradiation furnace calibration. This last test was conducted by placing the irradiated boiling fuel thermocouple rod (VA-2), together with a new thermocouple fuel rod (VA-3) and a chromel-alumel test thermocouple, in an argon-purged electric furnace located in a hot cell. A maximum temperature of 1696°F was reached with the arrangement. As shown in Table II, no obvious error pattern was followed by either fuel rod thermocouple.

Table II. Temperature Intercalibration between
W-W/26 w/o Re Thermocouple Rods
and Chromel/Alumel Thermocouples

Temperature (°F)		
Chromel- Alumel Thermocouple	Irradiated Fuel Rod VA-2	Nonirradiated Fuel Rod VA-3
75	87	85
202	87	85
297	305	283
420	438	426
512	530	511
589	610	595
693	708	695
800	815	808
915	925	920
1002	1011	1010
594	604	598
991	1002	998
1102	1103	1105
1192	1191	1195
1301	1300	1310
1400	1400	1405
1511	1505	1515
1600	1600	1610
1696	-	1705
1499	1498	1512
1301	1298	1315
1096	1088	1105
900	893	910
695	692	704
519	518	525

The only known errors worth consideration are the manufacturers' standards for the thermocouples. For the chromel-alumel thermocouple, this is $\pm 4^\circ\text{F}$ for temperatures to 530°F and $\pm 3/4\%$ of temperature for values greater than 530°F . Errors for the W-W/26% Re thermocouple are stated as $\pm 8^\circ\text{F}$ for temperatures up to 800°F and as $\pm 1\%$ of temperature for values above 800°F . Consideration of these errors alone leads to the conclusion that no measurable calibration change was produced in the W-W/26% Re thermocouple after a neutron-flux exposure of approximately 2×10^{18} nvt in BORAX-V.

3. Boiler Inlet Velocity and Exit Void Fraction Measurements with Core PSH-1

Measurements of the inlet velocity and exit void fraction were made with turbine-type flowmeters at the entrances and exits of instrumented boiling fuel assemblies I-1 and I-3. For example, Figure 5 shows inlet velocity as a function of reactor power for assembly I-1. Velocity is reported at the fuel assembly inlet nozzle, which has an effective cross-sectional area of $4.69 \times 10^{-2} \text{ ft}^2$ at the point of measurement by the turbine. Values of velocity were determined from the volumetric readings of the meters by applying the above area factor. No other correction factors were applied. The dual curve cannot be explained except by some abrupt change in each of the turbines which might be associated with the reactor shutdown. (A change in the friction of the turbine bearings is a possible explanation.) A post-operation calibration of the recorders associated with the meters showed no calibration errors greater than $1/2$ to 1% of full scale. (This would account for about 5% of actual velocity change at the flow range in question.) No changes in reactor operation which might account for the dual curves are known at this time.

Figure 5. Water Inlet Velocity to Instrumented Fuel Assembly I-1, from Turbine Meters, Core PSH-1 of BORAX-V

Exit void fraction measurements for I-1 are shown in Figure 6. The results shown were determined by entrance and exit turbine meter measurements and are void fraction values at the exit meters. Due to fluid expansion and contraction effects between the fuel exit and the meter, the actual fuel exit void fraction values might be expected to differ from the reported values by about 10% of value. (All exit void fraction values reported during BORAX-V operation are designated as volumetric percentage of steam in the exit mixture, measured at the exit turbine meter.)

Figure 6. Exit Void Fraction vs. Reactor Power for Fuel Assembly I-1, from Turbine Meters, Core PSH-1 of BORAX-V

4. Reactor Subcooling Measurements

Figure 7 summarizes the subcooling temperature measurements made with core PSH-1. Seven thermocouples were used to obtain the curve in the following manner: six thermocouples at the inlets to core positions 12, 21, and 22 measured water inlet temperature; a seventh thermocouple measured steam dome temperature. Periodic zero-power calibrations were made between the steam thermocouple and the six inlet thermocouples to account for random calibration changes, and all readings were then corrected on the basis of these calibrations. The six corrected

readings were then averaged for each data point and subtracted from the steam temperature at that time. The resulting average subcooling values were then plotted.

Figure 7. Reactor Subcooling vs. Thermal Power for Core PSH-1 of BORAX-V

Typical correction values ranged from $+1$ to -1°F , and points up the difficulty experienced in making the determinations of subcooling. Making accurate measurements of small temperature differentials remains as one of the problem regions of in-reactor vessel measurements.

5. Reactor Physics Calculations

Several 4-group PDQ problems have been solved for BORAX-V minimum critical experiments. These experiments, completed prior to loading the PSH-1 core, consisted of finding the minimum critical array of 12-flow-rod boiler assemblies and attempting to find a minimum critical array of 680-g superheater assemblies.

The minimum critical array of 12-flow-rod boiler assemblies consisted of twelve assemblies in a 3×4 array plus three rows of fuel rods and flow rods along the long side of the rectangle, giving a $3\frac{3}{7} \times 4$ array. The measured k_{eff} for this array with no poison rods was 1.0043 with the control rods at 25 in. and 1.0023 with two boron-stainless steel poison rods replacing fuel rods.

A critical array of 680-g superheater assemblies was not attained, since only 16 assemblies were available. However, the inverse count rate as a function of number of assemblies indicated that 18 assemblies would be supercritical.

The constants used in the PDQ calculations were obtained with the use of TEMPEST to obtain thermal constants (from 0 to 0.414 eV) for pin-cell (boiler) or plate-cell (superheater) calculations, and with GAM-I to obtain three fast-group constants.

The superheater plate cell flux distributions were obtained with the aid of FLIP-1, a double P_1 approximation, and the boiler pin-cell flux distributions were obtained from TUPIC, a cylindrical S_4 code with isotropic scattering outer boundary condition. The individual region constants (UO_2 , stainless steel clad, and water) were flux-weighted to give homogenized thermal constants for the boiler fuel zone or superheater fuel zone in PDQ.

The thermal constants for other material zones (water, aluminum-water mixtures, stainless steel, etc.) were averaged over the appropriate homogenized assembly spectrum.

A new U^{238} library (#139) has recently been added to the GAM-I library, and a comparison of the effect of fast constants using the old U^{238} (#12) cross sections with #139 on k_{eff} was made. The comparison of calculation with experiment is given in Table III.

A more detailed description of these problems and the methods used to obtain constants will be given in a final BORAX-V neutronics report.

Table III. Comparison of Calculated k_{eff} with
Minimum Critical Experiments

Configuration	Experimental k_{eff}	Calculated k_{eff}	PDQ Problem Number
Sixteen 680-g superheater assemblies	<1.000	1.0195 $\pm$ 0.0025	1001; stainless steel spacers between fuel plates omitted
		1.0100 $\pm$ 0.0025	1003; spacers included
12-flow-rod boiler minimum critical array, no poison rods	1.0043	1.0308 $\pm$ 0.0025	2001; using GAM #12 for U^{238}
		1.0060 $\pm$ 0.0025	2003; using GAM #139 for U^{238}
12-flow-rod boiler minimum critical array with 2 poison rods	1.0023	1.0032 $\pm$ 0.0004	2005; using GAM #139 for U^{238}

II. LIQUID-METAL-COOLED REACTORS

A. General Fast Reactor Physics

1. ZPR-III

Assembly 45 (see Progress Report for June 1964, ANL-6912, p. 16) has a multizoned core designed principally to investigate the Doppler effect in plutonium carbide-fueled power breeder reactors. For the first series of measurements, a portion of the sodium has been replaced with graphite to increase the magnitude of the Doppler effect and to provide an additional experimental value suitable for checking calculations.

A longitudinal cutaway view of the assembly for these measurements is given in Figure 8, showing the position of the Doppler pellets in the full "in" position. The Doppler pellets are encased in elements having a longitudinal cross section described in Figure 9. These elements are retracted axially to allow measurement of the dummy reference reactivity.

The dummy reference is composed of ZPR-III plates whose average composition and mass duplicate closely the true element. Exchange of the Doppler element for the dummy thus causes a very small reactivity difference which is measurable with a sensitive calibrated rod.

Figure 8. Doppler Assembly in ZPR-III-45

Notes

1. The inner surface of the outer can is silvered to prevent radiation losses. The space between outer can and sample can is evacuated. Cooling air is circulated outside the outer can.
2. The heater is an insulated resistance wire spirally wrapped around the sample can. The pitch of the spiral is varied to compensate for heat losses from the ends.
3. There are three thermocouples located on the center line, and one on the sample can of each sample.

Figure 9. Doppler Elements

Certain items bearing on the ultimate accuracy of reactivity measurements with available equipment have been investigated. For example, the position of the sensitive control rod is indicated by a selsyn in the control room. A dial indicator was used on the control rod itself to check the reading of the selsyn, which was found to be reproducible at three different positions within an average deviation of ± 0.00032 in. This corresponds to an error of about $\pm 0.05 \times 10^{-7}$ in reactivity.

Both temperature and humidity changes can affect reactivity slightly. Based on experience with other assemblies, a 0.01°C uniform change in temperature could change reactivity by $\sim 2 \times 10^{-7}$. A localized temperature change would have to be correspondingly larger to cause the same effect.

In spite of this extreme sensitivity, periods of up to several hours have been observed when the reactor was sufficiently stable, or drifted at a slow enough rate, that measurements could be made with a reproducibility of better than 10^{-7} . Data from two Doppler elements at 1200°K and corresponding data at 500°K have been used to form a preliminary estimate of 1.3×10^{-5} for the effect of heating the elements over this range. An example of the data is shown in Figure 10, which shows recent data taken with a single element at 500°K . The filled points represent critical positions with the dummy element in the core, read on the scale at the left; the open circles, representing critical positions with the Doppler element in the core, are to be read on the right-hand scale. The right and left scales are shifted to show the correspondence between the two types of measurements. The fine control rod is worth approximately 2×10^{-5} per in. The solid and dashed lines indicate the inferred drift of the reactor for "Doppler out" and "Doppler in" measurements, respectively. Over the period of time indicated by the lines, these data indicate a deviation of individual points less than 5×10^{-8} .

Figure 10
Worth of a Single Doppler
Element at 500°K
(Relative to a Dummy)

2. ZPR-VI

a. Assembly No. 3. Assembly No. 3 of ZPR-VI, a large (~ 950 -l), pancake shaped, uranium carbide-fueled core, is suitable for studying the sodium void coefficient because of large axial neutron leakage. The critical mass was 850 kg of U^{235} . An outline of the assembly and the rod positions are shown in Figure 11.

ZPR-VI
MASTER LOADING CHART

APPROVED BY _____

1. ASSEMBLY NO. _____
2. LOADING NO. _____
3. DATE _____

☒ D.P. CONTROL ROD
■ INSERTION SAFETY ROD

MOVABLE HALF
STATIONARY HALF HAS SAME LOADING

Figure 11. Loading of ZPR-VI Assembly No. 3

b. "Fat Man" Studies. Because of the unusual shape and extreme size of this assembly when fully loaded, it was of interest to determine the approximate degree of coupling when large amounts of moderating material were placed between the separated reactor halves. This was accomplished by determining the multiplication of each half for different sizes of "fat men." "Fat Man" measurements in which approximately 60 kg of hydrogen were placed between the reactor halves showed that the system was far from critical when the halves were separated.

The geometry for the coupling experiment is shown in Figure 12. Four counters and a source were used in the stationary half. In the stationary half, a Benelex encased BF_3 counter was located at the top of the core at its midplane and a U^{235} counter at the center of the core, but 2 in. back from the midplane. In the movable half, a second Benelex encased BF_3 counter was located at the top of the midplane and a He^3 counter in the core center, but 3 in. back from the midplane. The results of the measurement are shown in Figures 13 and 14.

Figure 12. Counting Geometry during "Fat Man" Effect Measurements.

Figure 13. Results of the "Fat Man" Experiment. Count Rates of the BF₃ Counters on Top of the Matrix.

Figure 14. Results of the "Fat Man" Experiments. Count Rates of Detectors in Center Drawers.

As the thickness of the Benelex was increased, the fission counter located in the center of the stationary half showed a strong increase in counting rate due to reflection and moderation, and the BF_3 counter located on top of the stationary half showed a strong decrease due to shielding.

The count rates of the He^3 counter in the center of the movable half and of the BF_3 counter on top of the stationary half showed similar characteristics because both of these counters were mostly oriented to see the movable half. The curves show that for the first 2 in. of thickness of the Benelex the shielding effect was offset by the moderation of the transmitted neutrons, which tended to increase the number of fissions and therefore the flux (increased coupling), whereas for larger thicknesses the shielding effect predominated.

c. Rod Calibration. The control rod calibrations were made by positive-period, rod-drop, and digital-rod-travel techniques. The measurements by the digital rod travel method¹ were taken both with falling and rising reactor power. The measurements at rising power had to be taken in two steps to avoid positive periods shorter than 30 sec.

Figure 15 shows a comparison of calibration curves of dual-purpose rod No. 1 found by the various methods. The fluctuations in the experimental data were not higher than expected from the counting statistics.

Figure 15
Calibration of
Dual-purpose
Rod No. 1

¹Reactor Development Program Progress Report, ANL-6860 (Feb 1964).

Total reactivity worths of several dual-purpose rods and boron safety rods are listed in Table IV.

Table IV. Total Reactivity Worths of Dual Purpose and Boron Safety Rods

	Period Measurement, Ih	Digital Method, Ih		Rod Drop, Ih
		Rising Power	Falling Power	
Dual Purpose Rods				
No. 1	89.2 ± 0.5	89.6 ± 0.8	90 ± 1.5	91 ± 3
No. 2	88.2 ± 0.5	87.1 ± 0.8	88 ± 1.5	-
Boron Rods				
No. 1				40.6 ± 2
No. 5				52.5 ± 4

d. Gap Worth. The measured gap worths as determined by calibrated control rods for air and stainless steel are plotted in Figure 16.

Figure 16
Gap Worth in ZPR-VI
Assembly No. 3

Measurements with air were made first with the tables opening and then with the tables closing. The gap width is not defined exactly because the surfaces of the reactor at the interface are not parallel. The average of the separation at the four interface corners was used as the effective gap width.

At large table separations, the gap worth was determined by the subcritical source-multiplication technique. Figure 17 shows the inverse counting rate versus table separation. The data from each counter were normalized to a reference run at a known subcritical, k_{ex} , so that they are equal to $1 - k_{eff}$ in the limit of small table separations, for which the effects of higher harmonics in the flux distribution can be negligible.

Figure 17
Inverse Multiplication
vs. Table Separation

Two entirely different neutron source distributions, a point source near the edge of the core and the evenly distributed source of the spontaneous fission neutrons, were used to test the effect of higher harmonics. The BF_3 counter used was sensitive to the source distribution for gap widths >0.5 cm, whereas the fission chamber was not sensitive to source distributions for gap widths less than about 10 cm.

e. Spatially Dependent Drawer and Fuel Worths. Measurements of the reactivity worths of a single drawer at the core center (S2323), 10 in. from the core center (S2328), and of four drawers at the edge of the core (average of drawer positions S0923, S2309, S2337, S3723) were made. The results are listed in Table V. In each case, the drawer was replaced by a void.

Table V. Drawer Reactivity Worths

<u>Drawer Position</u>	<u>Reactivity Worth/Drawer, lh</u>
S2323	42.7
S2328	34.5
S0923, S2309, S2337, S3723	6.1

Measurements of the reactivity worths of the U^{235} fuel columns were made relative to a void at the core center (S2323); relative to U^{238} at a point 4 in. from the core edge (average of four measurements at drawer positions S2311, S2335, S1123, S2523); and relative to U^{238} at the core edge (average of four measurements at drawer positions S0923, S2309, S2337, S3723). The results are given in Table VI.

Table VI. U^{235} Fuel Worths

<u>Drawer Position</u>	<u>Reactivity Worth/kg U^{235}, lh</u>
S2323	60.4
S2311, S2335, S1123, S2523	10.6
S0923, S2309, S2337, S3723	5.4

The reactivity worth of a fully loaded drawer was also measured relative to a blanket drawer at the core edge (positions 1232, 1412, 1434, 3212, 3234, 3414 in both halves). The measured worth of the twelve drawers was 32.8 lh or 2.73 lh per drawer.

B. General Fast Reactor Fuel Development

1. Metallic Fuels

Uranium-plutonium-base alloys are being investigated for use as fertile-fissile fuel materials in fast breeder reactors because many combinations of the base elements, which are readily available, have good neutron yield and breeding ratios. Alloying additions appear necessary,

however, to improve the stability of the uranium-plutonium base in a fast-reactor environment and the compatibility of the fertile-fissile alloy with jacketing materials. Fizzium (Fz), which is a relatively high-zirconium combination of desirable fission-product metals retained in the fuel on reprocessing, has good alloying possibilities. The properties and behavior of uranium-plutonium-fizzium (U-Pu-Fz) alloys, as well as various combinations of the constituent elements, are therefore being investigated.

a. Uranium-Plutonium-Fizzium Alloys.

(i) Transformation Kinetics of U-10 w/o Pu-10 w/o Fz Alloy.

The transformation kinetics of the potential fast reactor fuel U-10 w/o Pu-10 w/o Fz were studied by use of metallographic, X-ray diffraction, dilatometric, and tensile-test techniques. The results verify and supplement previously reported hardness-time curves.²

The transformations at 500 and 450°C are essentially complete after 30 min. The transformation, which is time dependent, results in a very fine microstructure. Overaging for one week results in a lamellar structure suggestive of a eutectoidal decomposition. The retained phase has an X-ray diffraction pattern resembling one that is body-centered cubic, but with a slight shift in lines from a pattern of a similar alloy not containing plutonium. The products of transformation are alpha uranium and gamma prime (U_2Mo structure), both containing plutonium.

The end of transformation was best detected by measurements of elastic modulus or hardness. Dilatometry was useful, although reaction of the specimen with the lead quenching medium confused the end point. Metallography was suitable for observing the initial and overaged structures, but a clear indication of the end point was not obtainable by this technique.

(ii) Creep Rate of the U-15 w/o Pu-10 w/o Fz Alloy. Because their tensile strength at elevated temperatures is low, the fizzium alloys ought to creep under their own weight. Creep tests of an injection-cast rod (0.365 cm in diameter by 5 cm long) were made with a dial-gage dilatometer. A compressive load of $11.6 (10^{-3}) \text{ kg/mm}^2$ was applied to the rod to account for the weight of an EBR-II fuel rod plus the weight of a blanket rod. Tests were at 650, 750, 850, and again at 650°C for about 170 hr at each temperature. After about 50 hr, the rod had contracted 0.04% at 650°C, 0.08% at 750°C, and 0.32% at 850°C. A fairly constant creep rate was obtained for the next 120 hr.

(iii) Compatibility of U-Pu-Fz Alloys with Potential Cladding Materials. The compatibility of U-15 w/o Pu-10 w/o Fz with V-20 w/o Ti

²Metallurgy Division Annual Report for 1963, ANL-6868.

has been evaluated in the temperature range from 600 to 750°C. A comparison of the data (see Figure 18) with those obtained for U-10 w/o Pu-10 w/o Fz versus V-20 w/o Ti (see Progress Report for July 1964, ANL-6923, p. 20, Figure 5) indicates, as expected, a somewhat accelerated cladding penetration due to the higher plutonium content. All the data can be adequately described by

$$X^2 = Kt$$

where X is the penetration into the cladding in centimeters, t the time in seconds, and K the penetration coefficient in cm^2/sec .

Figure 18
Penetration of V-20 w/o Ti by
U-15 w/o Pu-10 w/o Fz at Vari-
ous Times and Temperatures

No evidence for melting or accelerated penetration was detected at even the highest test temperature (750°C).

2. Carbide Fuel Elements

a. Production of PuC and (U, Pu)C₂ and Fabrication of Carbide Fuel Element Specimens. For fast breeder reactor solid-solution uranium-plutonium monocarbide fuels are usable at higher temperatures than are solid metallic fuels and are superior in thermal conductivity to the oxide fuels. Methods are being developed for producing vibratorily compacted carbide specimens for radiation studies in EBR-II. The fuel is solid-solution UC-20 w/o PuC with uranium enrichments of both 93 percent and 50 percent.

Raw materials, consisting of UC, 93 percent enriched in U²³⁵, depleted UC, and PuC containing 4.7 w/o carbon, were weighed, physically mixed, and micropulverized. The minus 44-μ fraction, 97.5 percent of the total, was mixed with a 0.5 w/o Carbowax 5000 binder. Green pellets were

pressed at 8.5 te/cm^2 (60 tsi), crushed, and then sized to 20/30 mesh (U. S. Standard). These granules were sintered in a helium atmosphere at 1800°C for one hour. Weight loss during sintering was negligible, and the molybdenum crucible showed little signs of attack. The sintered cake was easily granulated; only two percent of the particles were smaller than 30 mesh. Carbon content of the charged material was calculated at 4.78 w/o, but the product analyzed 4.62 w/o. Density measurements by immersion in bromobenzene gave 11.9 gm/cc , about 88 percent of theoretical. Attempts to increase the density will be made by refiring.

3. Development of Jacket Materials

a. Vanadium Alloys

(i) Creep of V-20 w/o Ti. Data for minimum strain rate versus stress at 550°C have been generated from one recrystallized specimen. The data were taken at strains from 0.12 to 0.20, and the stress levels were corrected to compensate for the reduction of area at these high strains. A comparison is made in Figure 19 with data previously reported (see Progress Report for July 1964, ANL-6923, p. 23) for stress-relieved and recrystallized specimens tested at 650°C . The curves are nearly parallel and have a slope of around $0.23 \text{ mm}^2 \text{ kg}^{-1}$. An apparent activation energy calculated for the recrystallized condition is 53 kcal/g-atom . The minimum creep rate of recrystallized V-20 w/o Ti in the testing range can be approximately expressed as a function of stress and temperature by the relationship

$$\dot{\epsilon} = 1.3 \times 10^3 \exp(0.23\sigma) \exp(-53,000/RT)$$

Figure 19
Creep of V-20 w/o Ti
at 550 and 650°C

where

$\dot{\epsilon}$ = minimum strain rate, sec^{-1} ,

σ = stress, kg mm^{-2}

R = gas constant, $\text{cal (g-atom)}^{-1}(\text{°K})^{-1}$

T = temperature, °K

(ii) Fabrication of Tubing. Tube blanks of vanadium-titanium alloy measuring 1.91 cm OD x 1.27 cm ID (3/4 in. OD x 1/2 in. ID) have been shipped to the Superior Tube Company and the Wolverine Tube Company for evaluation of commercial tube fabrication. A piece of the same size was sent to Fansteel Metallurgical Corporation for tube fabrication as their extrusion attempt did not yield an acceptable tube blank for further processing.

(iii) Alloy Development. In an effort to enhance the creep properties of the vanadium-titanium alloy, a series of vanadium-titanium alloys with varying additions of chromium are being prepared for study. Vanadium-titanium alloys containing molybdenum, tantalum, and/or niobium are also being prepared.

To date, two alloys (V-15 w/o Ti-5 w/o Cr and V-15 w/o Ti-7.5 w/o Cr) have been cast and are being prepared for rolling. The material for the other alloys to be made has been weighed and is now in the process of being arc melted.

4. Irradiation of Fast Reactor Fuels

a. Development and Fabrication of Irradiation Test Specimens. In a program for the development of fuel elements for fast breeder reactors, uranium-base fuel alloys containing 10 to 20 w/o Pu and 10 w/o Fz are being prepared for irradiation testing in EBR-II. A total of 97 test fuel pins have been cast, gamma radiographed, and machined to length. An additional 27 castings have been made.

Nineteen EBR-II fuel-rod irradiation test capsules are ready for reactor test. The fuel alloy and cladding material in these capsules are shown in Table VII. The EBR-II test capsule is a fairly complex fabrication. It consists of the fuel pin, a sodium bond, the fuel-rod jacket, a second sodium bond, a vanadium barrier thimble that is open at the top, a third sodium bond, and the outer stainless steel jacket. A capsule-loading sequence has been developed that greatly reduces voids and gas bubbles in the various sodium bonds. Measurements of capsule sodium levels from radiographs made at 25°C were very irregular and unreliable because of the shrinkage voids created in the sodium annuli during solidification. Radiographs at 150°C for 16 of the capsules showed sodium levels within

the specified range of 2.54 ± 0.32 cm (1 ± 0.125 in.) above the top of the vanadium thimble. One capsule showed a sodium level below the specified minimum and two capsules showed sodium levels above the maximum. Although some voids could be seen in the sodium annuli, it was not felt that they were sufficiently well resolved for dependence upon radiography for void determination. The capsules were then leak detected, surveyed for contamination, and inspected for conformity to the drawings.

Table VII. Fuel Composition and Jacket of Test Fuel Rods

Capsule Identification	Fuel Alloy	Jacket
C-93, C-97, C-98, C-99, C-100, C-101, CM-01	U-20 w/o Pu-10 w/o Fz	Nb-1 w/o Zr
CA-01, CB-02, CB-03, CB-04, CD-01, CD-02	U-15 w/o Pu-10 w/o Fz (U 7.7% enriched)	Nb-1 w/o Zr
LA-02	U-15 w/o Pu-10 w/o Fz (U 7.7% enriched)	Hastelloy-X with W Liner
PA-01		Nb-4 w/o V
CG-02, CG-03, CJ-01, PB-02	U-10 w/o Pu-10 w/o Fz (U 15.5% enriched)	Nb-1 w/o Zr

*Tungsten-electrode inert-gas arc.

b. Irradiation of Fast Reactor Metallic Fuels Postirradiation examinations were conducted on six U-20 w/o Pu-10 w/o Fz refractory alloy-jacketed specimens from capsule CP-35 irradiated in CP-5 (see Progress Report for July 1964, ANL-6923, p. 26). The specimens had been irradiated at maximum clad temperatures of 635°C for a calculated 2.5 a/o burnup. The rods initially measured 85 mm in length and 4.4 mm in diameter. The jacket material was Nb-1 w/o Zr, having a wall thickness of 0.23 mm. Three of the jackets were in a stress-relieved condition while the other three were in a fully recrystallized condition. The fuel pins measured 50.8 mm in length and were sodium bonded to the jacket. Four of the pins measured 3.66 mm in diameter and had 1.4 mm D axial holes. The other two measured 3.35 mm in diameter. Both configurations of fuel pins represented comparable fuel volumes. The jackets of the specimens containing the reduced diameter pins were purposely vented to the irradiation capsule environment. A thermocouple well was introduced into the axial hole of one specimen and central temperatures were read directly.

When the specimens had attained a calculated 1.2 a/o burnup, the capsule was removed from the reactor and neutron radiographed. From the radiograph it was possible to observe dimensional changes of the fuel pins. All of the pins having the axial hole, with the exception of

the one with the thermocouple, showed a reduction in length. The axial hole had collapsed in the midsection of the pin, and tapered gradually to both ends. The pins having the smallest visible existing hole openings had slumped the most, i.e., 6%. Conversely, the pin showing the largest area of opening remaining from the original hole had only a 2% reduction in length. The specimen containing the thermocouple revealed that the portion of the well immediately above the fuel was damaged and that the specimen was now vented to the capsule environment. The axial hole below the thermocouple well was not visible. This particular pin had elongated 4%. The two specimens that were fueled with 3.35-mm-diameter pins had elongated an average of 9.5 percent. The jackets of all specimens appeared intact and the capsule was returned to the reactor.

When the specimens had attained a calculated 2.5 a/o burnup the instrumenting thermocouples indicated variations suggestive of jacket failures. The jacketed specimens were removed from the reactor and, by neutron radiography, it was possible to see jacket deformations of four specimens. Disassembly of the capsule revealed jacket failures of five specimens and fuel protruding from the vent hole of each of the two vented specimens. Measurements taken from radiographs revealed that two of the specimens having axial holes still showed a 6% reduction in fuel length. The other fuel pin with the axial hole had slumped an additional percent. The pin with the thermocouple well had elongated one percent. The axial holes were no longer visible except at the extreme ends of the pins. Part of the thermocouple well, however, could still be seen. The two vented pins (originally of 3.35-mm diameter) retained the 9.5% average value for fuel elongation.

Dimensional measurements of the rods showed no appreciable length changes. The diameter of the rods remained the same with the exception of the areas affected by the protrusion of the fuel material. The diameter measurements taken at right angles to these areas of failure showed an average of 0.3% increase.

The location of the failures coincided with the calculated highest-temperature position of the rod. The appearance of the fuel material emerging from the jackets of three specimens is characteristic of unrestrained swelling of this fuel. The appearance of the fuel material protruding from the other three specimens indicates that at one time or another the fuel might have approached a liquid state. Metallographic studies, which will be conducted on selected sections of these rods, will assist in clarifying this question.

The two configurations of fuel pins, i.e., axial hole and smaller diameter, originally represented comparable fuel volumes. Presuming that expansion of the fuel was at a uniform rate for both configurations it would be anticipated that fuel material would completely fill the jackets

radially at approximately the same time. Observations of the size and type of jacket failures indicate that the failures did occur at about the same time. The only intact specimen was a small-diameter pin that had been jacketed in vented tubing that had been fully recrystallized. The companion specimen, which was jacketed in vented tubing in a stress-relieved condition, had failed. Three of the jackets had originally been in a fully recrystallized condition, but the appearance of the failed sections indicated brittle fracture of the material. The nature of the failures precluded direct comparison with jackets that were originally in the stress-relieved condition.

The estimated burnup figures reported for these specimens will be checked experimentally by mass spectrometer isotopic analysis.

Capsule CP-38 is presently being irradiated in the CP-5 reactor. The capsule contains six pins of U-15 w/o Pu-10 w/o Fz that are jacketed in either Type 304 stainless steel or Hastelloy X. The fuel is separated from the 304 stainless steel jackets by a vanadium liner and from the Hastelloy X jackets by vapor-deposited tungsten. The jacket ID's are either 3.96 or 4.2 mm, and the wall thicknesses 0.23 or 0.38 mm. Three of the specimen jackets have been vented to the capsule environment. The jacket temperatures are being maintained at 550°C and the irradiation period will be extended until there is evidence that jacket failure has occurred.

Six fuel specimens jacketed in V-20 w/o Ti tubing are being irradiated in capsule CP-40. The rods are fueled with U-15 w/o Pu-10 w/o Fz pins that measure 3.66 mm in diameter and 50.8 mm in length. The wall thickness of all tubes is 0.4 mm, but the ID's are either 4.0, 4.1, 4.2, or 4.3 mm. The internal dimensions of each rod were carefully controlled so that at a calculated fuel burnup of 2.5 a/o the jackets should fail. Axial fuel restraint is provided by piston type plungers spaced a predetermined distance from the top of the fuel. The specimens are being irradiated at jacket temperatures of 600°C.

c. Irradiation in Gas-annulus Capsules. Prototype fuel rods for EBR-II are being irradiated in the MTR in gas-annulus capsules.

Capsule ANL-55-8 contains three sodium-bonded fuel rods as follows:

U-15 w/o Pu-10 w/o Fz clad with Nb-1 w/o Zr;

U-10 w/o Pu-10 w/o Fz clad with Nb-1 w/o Zr;

U-15 w/o Pu-10 w/o Fz clad with vanadium.

The uranium in the 15 w/o Pu alloy is 7.7% enriched and that in the 10 w/o Pu alloy 15.5% enriched. The fuel pins are 33.02 cm long and

3.66 mm in diameter. The cladding materials were initially in an annealed condition. The ID of all cladding was 3.96 mm and the wall thickness was 0.38 mm. The average hot-spot cladding temperature has been about 555°C. Burnup is estimated at 0.5 a/o.

Capsule ANL-55-7 also contains three fuel rods. The fuel pins are all U-15 w/o Pu-10 w/o Fz with 7.7% enriched uranium. The pins are 33.02 cm long and 3.66 mm in diameter. The cladding materials are as follows:

Nb-1 w/o Zr, stress-relieved, 3.96 mm in ID, 0.38-mm wall;

Vanadium, stress-relieved, 3.96 mm in ID, 0.38-mm wall;

Hastelloy X with a tungsten liner, 3.88 mm in ID, 0.23 mm + 0.04 mm wall, respectively.

Sodium is the heat transfer medium between the fuel and the cladding. The average hot-spot cladding temperature has been about 555°C. Burnup is estimated at 1 a/o.

Capsule ANL-55-5 was removed from the MTR after an estimated 1.9 a/o burnup at an average hot-spot cladding temperature of 550°C. Thermocouples had given an indication of cladding failure. This capsule contains one fuel pin of U-20 w/o Pu-10 w/o Fs clad in Nb-1 w/o Zr tubing with 3.96-mm ID and 0.23-mm wall thickness. The fuel is sodium bonded to the cladding. The fuel pin was initially 36.1 cm long and 3.66 mm in diameter. A neutron radiograph taken of the encapsulated fuel rod after irradiation showed two areas of cladding failure accompanied by drastic swelling of the fuel. Both areas of failure were on the upper third of the length of the fuel pin and were about 3.2 cm (1.25 in.) apart. The uppermost failure was about 2 cm (0.75 in.) from the end of the fuel pin.

Capsule ANL-55-6 was removed from MTR after an estimated burnup of 1.9 a/o. The average hot-spot cladding temperature was 505°C. The temperature data for this experiment gave no evidence of any kind of failure. The three fuel pins in this capsule are U-20 w/o Pu-10 w/o Fs. They were 36.1 cm long and 3.66 mm in diameter. The three pins were clad in stress-relieved Nb-1 w/o Zr tubing with 3.96-mm ID and 0.23-mm wall thickness. The fuel pins were all sodium bonded. This capsule is still at the MTR waiting for shipment to ANL, Illinois.

5. Zero-power Reactor Fuels

a. Fabrication of Doppler Test Elements. Thirty-two test capsules are needed for study of the relationship between Doppler effect and plutonia concentration in ZPR-III. Half of the capsules are to be loaded with PuO₂ pellets, and the other half with pellets of UO₂-12.5 w/o PuO₂. Twelve cored

pellets, 1.27 cm in OD by 1.27 cm long, each with a 3.6-mm axial hole, are to be loaded into each capsule over a thermocouple protective tube. The outside of every capsule is covered by a spirally wound, brazed-in-place heating element. Before acceptance, a helium mass spectrometer is used to leak test each capsule at room temperature and at 1000°C.

Since the last reporting period, all the capsules for which hardware was provided have been loaded and welded closed. Of the sixteen capsules loaded with mixed $\text{UO}_2\text{-PuO}_2$, nine were found to be leak-tight to helium mass-spectrometer levels at 1000°C, four gave helium signals at temperatures below 600°C, one gave a slight leak indication at 1000°C, one showed current leakage from the heater wire to the capsule sheath, and one had a broken lead wire. Since all capsules were decontaminated to show less than 200-dpm direct count and zero wipe count, and since no increase in these levels was detected after testing, the capsules that indicated leaks at temperatures below 1000°C were considered usable for room temperature experiments.

Twenty-one capsules were provided for the loading of PuO_2 pellets. Five of the capsules were rejected for physical defects or leaks before loading. Of the sixteen remaining, three were scrapped in process because of interferences between pellets, loading funnel and capsule. Thirteen capsules were produced and to date five have been tested and found to be leak-tight at 1000°C.

C. General Fast Reactor Fuel Reprocessing Development

1. Skull Reclamation Process

The processing of oxidized skulls from melt-refining crucibles is being studied on a scale of 1.5 kg of uranium. In a recent run, a 10-in.-dia by 20-in.-high tungsten crucible that had been fabricated by welding was employed. Initial indications are that the crucible performed well. Analytical results have been received for the first run in which a reaction temperature of 700°C (instead of 800°C) was used for the noble metal extraction, reduction, and intermetallic decomposition steps (see Progress Report for June 1964, ANL-6912, pp. 29-30). Removal of fission products was satisfactory. About 87.5% of the uranium-zinc-magnesium product solution, containing about 84.0% of the total uranium charged, was transferred out of the tungsten process crucible at the end of the run. A uranium material balance of about 96.5% was realized in this run.

2. Consolidation of Fission Product Wastes in Alloys

Consolidation of fission products into metallic alloys appears to offer potential advantages both for the storage of radioactive wastes and for possible utilization of fission product decay heat. For these purposes, alloys of copper, aluminum, magnesium, and certain other metals have

the desirable properties of low cost, availability, satisfactory melting points, good thermal conductivity, corrosion resistance, and solubility of fission products. One experiment has been conducted to determine the distribution behavior of praseodymium between 30 m/o NaCl-20 m/o KCl-50 m/o MgCl_2 and aluminum-magnesium alloys at 600°C. The results are highly encouraging in showing aluminum-magnesium alloy to be an excellent reductant for rare earth fission products.

3. Decladding Studies for TV-20 Cladding

Several methods of separating vanadium-20 w/o titanium (TV-20) cladding from metallic fuels for pyrometallurgical processing are being investigated. In an earlier experiment (see Progress Report for July 1964, ANL-6923, p. 30) in which uranium and TV-20 were chlorinated in 40 m/o NaCl-30 m/o KCl-30 m/o CaCl_2 at 600°C, more than 99% of the uranium was retained in the salt and only 0.2% of the vanadium and titanium was retained. In a recent experiment, pieces of TV-20 tubing and uranium-fissium alloy rod were chlorinated on a gram-scale in quartz apparatus to determine the behavior of the fissium elements. The alloy pieces were immersed in 40 m/o NaCl-30 m/o KCl-30 m/o CaCl_2 and were chlorinated at 600°C for about 45 min. About one-fourth of the uranium, most of the molybdenum, ruthenium, and palladium, and some of the zirconium were unreacted at the end of chlorination, suggesting that the presence of the fissium elements inhibits the chlorination of uranium-fissium alloy. About 99.6% of the TV-20 constituents were volatilized, as was about 1% of the uranium. Although the chlorination process is considered chemically feasible, the corrosiveness of the system and the necessity for handling relatively large volumes of gases are disadvantages.

A small-scale scouting experiment was conducted on the removal of uranium from TV-20 alloy by leaching with cadmium. A uranium rod encased in an open-end TV-20 tube was leached with cadmium in a quartz Soxhlet-type device. Nearly all of the uranium was dissolved at about 500°C, with no visible attack on the TV-20 tubing. A stainless steel extractor is being constructed for future experiments.

Another possible method of removing TV-20 cladding from metallic fuel is by hydriding and dehydriding the fuel alloy to form a powder that can be mechanically separated from chopped sections of cladding. In a scouting experiment, a uranium-fissium rod was hydrided under 2.5 atm of hydrogen at 250°C for about 3 hr and then was dehydrided under vacuum at 300°C for 1 hr. The product was a mixture of powder and particles up to 2 mm in diameter.

D. Sodium Coolant Chemistry

1. Analysis for Oxygen

a. Distillation Analyzer. Operation of the distillation analyzer provided with quartz observation windows (see Progress Report for August 1964, ANL-6936, p. 27) indicated that the height of the sodium meniscus after filling the cup, and consequently the volume of sodium in the cup, was affected by the presence of a nonwetable film deposited around the rim of the cup. The presence of the film at this critical region was believed due to the nearness of the annulus between the column supporting the cup and the surrounding baffle tube.

In a new analyzer being designed, the annulus will be located at a position some distance below the rim of the cup to minimize the deposition in this area.

2. Cover-gas Impurities

One of the sources of impurities in sodium used as a coolant in nuclear reactors is the cover gas which may contain contaminants absorbable by the sodium. To obtain a qualitative appraisal of the importance of this source, an experiment has been performed in which helium containing typical gas impurities was bubbled through liquid sodium at 300°C. The helium contained about 2000 ppm of each of the following gases: hydrogen, methane, nitrogen, carbon monoxide, oxygen, and carbon dioxide. After 1, 2, and 4 hr of sparging, the circulating gas was analyzed by mass spectrometry.

At the end of 2 hr, the concentrations of oxygen and carbon dioxide were each less than 100 ppm. The final methane concentration was approximately 1800 ppm. The hydrogen concentration dropped to about 1000 ppm in 2 hr and remained constant for the remaining 2 hr. Results for nitrogen and carbon monoxide indicate some absorption of carbon monoxide.

E. EBR-II

1. Reactor Plant

Plant maintenance and repair activities were carried out during this reporting period. The principal job was repair of the main fuel gripper, which had been damaged during the preparation for installation of a replacement oscillator rod in the reactor.

a. Damage and Repair of the Main Fuel Gripper. During preparations for installation of Oscillator Rod No. 2 in the reactor, the funnel of the main gripper contacted the holddown as the gripper was being lowered

at a speed of 6 in./min. Normally, the gripper will pass through the central hole of the holddown without interference. It is deduced that the gripper shaft became misaligned as the result of distortion caused by temperature changes in the shaft as it was being lowered in the sodium. It had been previously maintained in the "up," cool position for considerable time before it was lowered.

To determine whether the gripper was still in satisfactory condition, a transfer of the boron control subassembly from the storage basket to the control rod thimble vacated by the first oscillator rod, thence back to the storage basket, was attempted. Because proper insertion of the subassembly top adapter into the damaged gripper could not be made, the plan could not be carried out.

In a further test, the gripper was engaged with the top adapter of the inner blanket subassembly which had been transferred to the storage basket. The gripper would not accept this top adapter easily, and considerable force was required to obtain full insertion of the top adapter in the gripper jaws.

Following the tests described above, a subassembly top adapter having a slightly undersize "mushroom" was fastened in the transfer arm and used as a gauge to check the gripper funnel and jaw assembly. Engagement of this adapter with the gripper was difficult; and, after removal, the adapter "mushroom" was found to be deeply scored on one side, indicating probable distortion of the gripper funnel and jaw assembly.

No significant improvement in gripper-adapter alignment was found by slightly changing the transfer point through changes in rotating plug orientation.

Since the gripper has been damaged so that it is no longer usable, the plant coolant systems were cooled to about 320°F preparatory to removing the gripper mechanism from the primary tank. Spare parts were procured for replacement of the funnel and jaw assembly.

After removal of the gripper from the primary tank, examination of the jaw assembly showed that the guide funnel had been twisted about its support so as to rotate the funnel approximately 6°. This displacement was sufficient to prevent complete closure of the gripper jaws.

Gamma radiation at 2 in. from the bottom end of the gripper was about 6 R/hr. Therefore, shielding was required during disassembly and repair.

The drive shaft was cleaned with ethyl alcohol and water, and then vacuum-dried. A satisfactory helium leak test was performed on the internal bellows.

The gripper mechanism was replaced in the primary tank and plant heatup was started. When the primary sodium temperature reached 580°F, the gripper and other fuel-handling components were checked. During preliminary checkout of the gripper, it was found that complete opening of the gripper jaws was not possible with the electrical drive, and the sensing rod could not be moved. Soaking in the hot sodium and manipulation of the gripper mechanism led to freer movement of the gripper jaws and sensing rod. It is believed that sodium oxide deposits were responsible for the initial difficulties. As the report period ended, the gripper was restored to operative condition, and checkout of the fuel-handling systems was proceeding. Checkout of the oscillator drive mechanism was also in progress.

b. Inspection of Primary Sodium Pump Motors. The drive motors of the primary sodium pumps were partially disassembled in place (without being removed from the pump shafts) to permit investigation of the cause of oil loss from the reservoir which supplies lubricant to the top thrust bearing of the motor. This loss had been observed in both pump motors during prior operation, and excessive oil consumption was confirmed by reviewing maintenance records.

Examination of the interior of each motor after removal of the thrust bearing and the top cover of the motor case showed no visible extensive oil deposition on the motor shaft, top gas flow baffle, or the visible metallic parts of the rotor. There was a thin film of oil on the underside of the motor top cover, that appeared to have come through a gas pressure-equalizing pipe which communicates with the oil reservoir. There also seemed to be a very thin, uniform film of oily substance on the stator windings accessible to inspection.

However, in each pump motor, about $2\frac{1}{2}$ quarts of oil were found in a welded "box" adjacent to the oil reservoir, which forms part of a structure through which the power leads enter the pump case. It communicates with the oil reservoir via a small hole near the top of the box through which pass the electrical leads from the tachometer located immediately above the oil reservoir. Oil apparently had been thrown or splashed through this small hole by the action of the bearing, particularly at high pump speeds. Once the oil entered the box, it could not return to the reservoir until the level in the box reached the entrance hole.

The quantity of oil found was sufficient to account for the observed losses from the oil reservoir. It is believed that only a very small amount of oil, probably as a fine mist entrained in the argon coolant gas, actually entered the motor case; this probably accounts for the oily film on the stator windings.

The top end of the gas equalizing pipe between the motor case and the oil reservoir was provided with a more effective shield to reduce the probability of splashed oil droplets entering the pipe.

With the apparent loss of oil from the reservoir adequately explained, the pump motors were reassembled. During assembly, the welded "box" described above was filled with oil to the level of the hole for the tachometer leads. Thus, this box no longer will act as a trap for the oil splashed from the normal reservoir.

c. Sodium and Argon. Primary sodium plugging temperatures were found to be less than 220°F.

The distillation technique for determining oxide content in sodium has been unreliable as used here under field conditions. Approximately 50 determinations have been made since April 1963, and in no instance has a distillation determination approximated the plugging meter result. Personnel of the Reactor Engineering Division are currently attempting to determine the shortcomings of the sampling and distillation method and to improve it for field use.

No further distillation samples will be taken from the EBR-II primary and secondary systems until the reliability of the method has been improved.

Gas chromatographic analyses of the primary argon showed the hydrogen concentration to be consistently less than 20 ppm by volume. Reported nitrogen concentrations varied between 0.3 and 1.7 v/o with the bulk of the values between 1.0 and 1.3 v/o. It appears that during the latter half of the month the nitrogen concentration has leveled off at about 1.0 v/o.

The secondary sodium was dumped to the storage tank after the plant had been cooled to 320°F (see Sect. B1a above). It remained in the storage tank until September 18 when the system was refilled during the course of plant heatup to 580°F.

Prior to dumping of the secondary sodium, the continuous chromatograph in the secondary argon system indicated about 1000 ppm by volume of hydrogen-helium mixture and 500 ppm of nitrogen.

Before the secondary system was refilled with sodium, the system was evacuated and filled with fresh argon. The hydrogen-helium content (determined by analysis) was reduced to less than 20 ppm.

During the month, the plugging temperature of the secondary sodium ranged between 215°F and 300°F. The new permanent cold trap was placed in operation during refilling of the system.

d. Maintenance and Modification Work during Plant Shutdown.
In addition to the work on the main gripper and the primary sodium pumps, reported above, the following have been completed or are nearly complete at the end of the report period:

(i) A motorized operator was installed on the syphon break valve for the primary sodium purification system. This installation will permit automatic shutdown of the purification system in case of a sodium leak in this system.

(ii) The temporary secondary sodium cold trap and mixing line were removed.

(iii) The cooling coils of secondary sodium recirculating pump No. 2 were cleaned of debris from the cooling water system.

(iv) Block and bypass valves were installed in the cooling water lines of the sodium boiler plant to permit installation of strainers, when received, without necessity of a system shutdown.

(v) Provisions were made for installation of an improved vacuum control system for the main secondary sodium pump case (when components are available) without shutting down the system.

(vi) Installation of a "charged wire" fission gas monitor, similar to that which performed satisfactorily at EBR-I,³ was nearly completed.

(vii) Repair of the damaged expansion joint bellows, in the desuperheater line to the condenser, was completed, and the bellows was reinstalled in the line with minor modifications of the expansion joint to limit compression of the bellows under atmospheric pressure load when a partial vacuum exists in the bypass line at low steam flows. Provisions have been made for measuring the movement of the pipe tee within the expansion joint under steam flow conditions and also for measuring movement of the bypass line upstream of its anchor point.

(viii) The large steam bypass control valve was inspected and found to be in satisfactory condition. Upon the manufacturer's recommendation, new trim was ordered for the small steam bypass valve which performed unsatisfactorily during the power demonstration run because of extreme wear on the valve trim. At the end of the report period, the new trim had not yet been received; therefore, the small valve was still inoperative.

(ix) The turbine nonreturn valve installation was completed, requiring only operating adjustments to be made during the next power operation.

(x) Maintenance of instruments and controls in the Power Plant proceeded during the plant shutdown. All gauges were calibrated.

³Smith, R. R. and Doe, C. B., Fission Product Monitoring in EBR-I, Mark IV, ANL-6788 (Jan 1964).

A turbine shell temperature indicator was installed. An alarm for low sealing steam pressure was installed also.

(xi) An oil leak of the turbine initial pressure governor was repaired. Water and steam leaks throughout the plant, on heaters, piping connections, sight glasses, etc., were repaired. Manual valves were maintained and reconditioned, as time and manpower availability permitted.

(xii) The steam drum safety valves were removed, and the discs and seats were lapped. They have been set cold (by bench testing) to relieve at about 1390 and 1420 psig. It is expected that these settings will be checked hot and adjusted as necessary when a sufficiently high plant temperature is reached.

2. Reactor Physics

a. Antimony-Beryllium Source Regeneration. The demonstration run has increased the shutdown count rate on Channels 1, 2, and 3 by about 63 cps. Comparison of the original source irradiation history in MTR with the corresponding irradiation history in EBR-II together with proper allowances for decay and relative fast and thermal cross sections shows that this increase in count rate is in agreement with calculations. The decay rate of the present source has been followed since shutdown and indicates an approximate 64-day half-life. The possible contribution to the source strength due to gamma-induced neutrons in the beryllium is also being investigated.

b. Na²⁴ and Na²² Activities. The buildup of Na²² from the (n,2n) reaction has caused some concern with respect to future maintenance and repair activities. A very preliminary estimate of the buildup of this isotope was made from determination of the activities of Na²² and Na²⁴ measured by a pulse-height analyzer. After appropriate corrections for the decay of Na²⁴, the equilibrium Na²⁴ activity is calculated to be 800 $\mu\text{C/g}$ of sodium at 62 MW. From the Na²² activity, a relative cross section for these two isotopes was determined as

$$\frac{\sigma(22)}{\sigma(24)} = 3.2 \times 10^{-4}.$$

3. Fuel Cycle Facility

a. Installation and Testing of Fuel-refabrication Equipment. The status of this program is as follows:

(i) Charge-preparation Equipment. The 20-kg, remotely controlled weighing balance and its out-of-cell components have exceeded their specifications in sensitivity and accuracy. This item is ready for production operation. The pallet loading and unloading manipulator operations are also working satisfactorily.

(ii) Injection Casting. Both injection casting furnaces have been tested and are ready for production. Although the present dipping thermocouple with a coated tantalum tube functions satisfactorily if periodically replaced, an improved thermocouple protective tube is still being sought.

(iii) Crucible Skull Dumper. The crucible skull dumper operates satisfactorily.

(iv) Skull Breaker. The casting skull breaker is operational.

(v) Pin-process Machines. One pin-process machine under the master-slave manipulator at Cell Window 5 is operation. About 35 castings per hour can be demolded, sheared, and inspected.

The second pin-process machine is out of the cell for replacement of the gravity feed-type demolder feeder tray with a more positive mechanical feeder. The new demolder feeding tray has yet to be tested.

One data process unit has been modified with the installation of an encoder that is connected to an automatic IBM card punch machine. Other modifications increase the flexibility of the unit. The data processor for the second machine will also be reworked to match the first. The preparation of the second pin-process and inspection machine and its data unit will not delay start up of the process since these units were provided for emergency standby operation.

(vi) Sodium-loading Equipment. The sodium extruder and fuel-tube-loading equipment is operational.

(vii) Fuel-rod-assembly Equipment. The master-slave fuel-rod assembly operation has been tested and is ready for production.

(viii) Settling Machines. The settling machines are working satisfactorily and are ready for production operations.

(ix) Welding. The fuel-rod welder has been modified. A device has been added that positions the top of the fuel rod in relation to the welding electrode. The change from helium to argon shroud gas has increased the number of reject welds. This appears to be due partly to the increased power required to make satisfactory welds in argon, and partly to the decreased cooling rate of the weld bead. The resulting weld bead tends to sag if sufficient power is applied for complete fusion of the top of the fuel element. If the power is cut back to produce a smaller weld bead, fusion is incomplete and defective fuel elements occur. The sagging or mushroomed weld beads do not enter the leak detector heads, which are 4.6 mm (0.180 in.) in diameter.

(x) Leak Detectors. The leak detectors function properly on properly dimensioned welds and are ready for production.

(xi) Bonders. One of two bonders has been converted for pneumatic operation with the temporary installation of a nonradiation-resistant cylinder. This bonder functions satisfactorily, producing acceptable fuel rods in about 1000 impacts. The final modification of both bonders awaits the delivery of radiation-resistant pneumatic cylinders. These cylinder assemblies will be plug-in replacement for the present electrical drives. The pneumatic controls for the pneumatic actuators have been designed, constructed, and tested. The initial production can be handled with the present pneumatic bonder.

(xii) Bond Testers. Both the in-cell and external equipment for the sodium bond-and-level determination have been tested and are ready for operation. The difficulty previously reported with the lava coil form was traced to improperly fired lava. New coil forms have been provided that have been fired to full hardness.

(xiii) Fuel-element-assembly Machine. The 61-rod assembly equipment is ready for operation. A control rod was successfully assembled by remote means. The element was tested for straightness and pulled in tension. It was found acceptable, this being the first element made in the Fuel Cycle Facility that can be delivered to the reactor. Similar tests are under way on the 91-fuel-rod-assembly equipment.

(xiv) Straightness-and Tensile-testing Equipment. This equipment operates satisfactorily.

(xv) Grapples. The fuel-element grapples have been tested and appear to be operational.

(xvi) Air Cell Transfer Grapple. The grapple used for transferring core subassemblies from the interbuilding coffin into the Air Cell is currently operable without the in-cell hose support unit and the hose storage unit, which have been fabricated and currently are being readied for shipment to Idaho.

(xvii) Core-subassembly Dismantler. A device to assist the mechanical master-slaves in removing the blanket rods is being fabricated. This unit uses a pneumatically powered wedge to shear the blanket elements from the support grid. The wedge is driven between the rows of the elements at the grid, applying pressure to shear one element at a time in one row, while using the next row as a reaction surface. The unit is to be equipped with two sizes of wedges to permit removal of upper and lower blanket, as well as the larger, full-length, inner and outer blanket, elements.

b. Fuel Examination and Recovery

The Fuel Cycle Facility has now received its first radioactive core subassembly and inner blanket subassembly from the EBR-II reactor, which were removed after approximately 0.1% burnup of the central core element. After about 10 days of cooling, the core subassembly was disassembled in the Air Cell, and the cladding was removed from the fuel pins in the Argon Cell. After samples were taken for burnup analysis, the remaining fuel material was chopped, charged to the melt refining crucible, and processed without incident. The pouring yield was 92.7% and two samples of this product are being analyzed. The skull remaining in the crucible was successfully oxidized and transferred to a storage container.

No problems were encountered in the sodium-removal step. The activity level of the sodium removed was low, as the reactor had been shut down since removal of the subassemblies from the reactor. Decay heat from the core subassembly (calculated to be about 380 W) masked the heat effects of the sodium reactions, indicating satisfactory performance of the oxidation-hydrolysis system.

Some difficulty was encountered in removing the inner blanket assembly from the transfer cask, as the upper pole piece was bent near the top end, and manual assistance in grappling was necessary. The core assembly was grappled and removed from the cask without trouble. Radiation from the core assembly when it entered the Air Cell amounted to about 10^5 R/hr at one ft.

The inner blanket assembly, which was removed from Row 6, could not be checked for straightness because of the bent pole piece. No deterioration of stainless or chrome surfaces was apparent. The assembly was dismantled and five of the 19 rods were checked for diameter with go-no go gauges. No changes from specifications were noted.

The core assembly, which had been removed from Row 2, met initial specifications for straightness when tested in the final assembly test machine, and no adverse changes were noted on exterior surfaces. It was dismantled satisfactorily and eleven fuel elements (constituting a diametrical row) were segregated for detailed examination. The surface condition of all eleven appeared good, with no evidence of hot spots or sodium leaks. Diameters were checked with a 0.1765 gauge, and all passed through, indicating the absence of significant fuel swelling above can ID. Bond and level tests indicated the presence of some minor voids and a small, statistical increase in sodium level. Three of the elements were decanned to provide samples for chemical analyses and burnup measurement. Data are not yet complete. Following completion of Air Cell work, active materials were removed, and the cell was surveyed and

thoroughly vacuumed. When re-entered on September 11, little contamination was found other than a few stainless steel chips in the dismantler.

The remaining 80 fuel elements from the core assembly were decanned and chopped for processing. Two pins were incompletely decanned, and small stub ends remain in the cladding. The 5.4-kg balance plus 3.2 kg of unirradiated injection casting scrap and unalloyed enriched uranium constituted a melt refining run charge. The batch was liquated 3 hr at 1400°C to produce an ingot weighing 7.95 kg (92.7% yield). The ingot has been sampled for analysis, but data are not yet available. The crucible residue was oxidized, and an oxide sample was also obtained. As anticipated, the decanning, melt refining, and oxidation operations resulted in noticeable activity in the sub-cells. This was associated principally with the argon circulation cooling box, the purification system filter, and the furnace off-gas system.

The search for more suitable bearing materials for the decanner and other inert atmosphere cell equipment was continued with tests at very low levels of oxygen and water vapor (3 and 1 ppm, respectively). These conditions resulted in the greatest wear yet experienced. Limited data obtained to date suggest the following conclusions:

- (i) There is no correlation between the rate of wear of carbon-base bearing materials with physical properties.
- (ii) Carbon graphites are better than fully graphitized materials, but all unimpregnated materials wear rapidly under the test conditions.
- (iii) Impregnation of graphite with silver, molybdenum disilicide, resins, and molten salts effectively reduce wear, but cadmium, copper, and Babbitt metal are ineffective.

c. Fuel Fabrication. An experimental injection casting run was completed on recycle enriched fissium scrap in an attempt to improve casting efficiency. Normally, argon is used as the pressurizing and quench (cooling) gas in this operation. The substitution of helium for argon was investigated with the aim of improving cooling efficiency to produce a more uniform group of castings with respect to the alpha-gamma transformation. Such uniformity can be expected to affect favorably both shearing and constancy of sodium level.

Of 56 molds utilized in the casting in which helium was used for cooling, 55 were filled and 50 acceptable rods were obtained. Of the 5 pins rejected, one was due to operator error in shearing, two were diameter rejects (cold shuts), and two were of short length. Excluding one reject because of error, the 93% yield looks very good on a preliminary basis, in comparison with an average 71% yield for the seven preceding batches. Investigation of the use of helium will be continued.

Initial work was begun on the testing and reassembly of the approximately 2300 elements recovered by disassembly of various special subassemblies no longer needed. A sampling (18 groups of 10, out of every 91) indicated that the number of bubbles and high sodium levels indicated by radiography was such as to require reinspection (eddy current bond and level) of all the elements. Of three groups of 91 processed to date, 73 to 87% have been accepted after partial or complete rebonding, and additional rebonding and testing of the remainder is underway. Two subassemblies were completed using the remote equipment with a small amount of assistance for direct observation and guidance. The first subassembly, a 61-pin control rod, was made up of newly fabricated elements. The second, a 91-pin core subassembly, used primarily elements reclaimed from disassembly operation.

The control rod assembly operation proceeded without incident, and tensile and straightness test results were satisfactory. During assembly of the core unit, one of the guide mechanisms was inadvertently omitted, and a slight distortion of the lower edge of the hexagonal tube resulted. This produced a poor-looking weld and failure to pass the straightness test, but the tensile test was acceptable. The subassembly was removed from the cell for correction of the welding and straightening.

d. Skull Reclamation Process. The retorting step of the skull reclamation process has been demonstrated in engineering-scale runs by the Chemical Engineering Division at Argonne (see Progress Reports for July and August 1964, ANL-6923, p. 40, and ANL-6936, pp. 33-35). In the retorting step, magnesium-zinc is distilled from the material resulting from the uranium dissolution step, after which the residual uranium is melted to form an ingot. All of the process steps prior to the retorting step are conducted in a tungsten crucible. The retorting step is conducted in a beryllia crucible.

Tungsten crucibles appear to be more reliable than beryllia crucibles and are presently available in plant-scale sizes, whereas beryllia crucibles are not. It would be advantageous if a tungsten crucible could be used in the retorting step instead of a beryllia crucible. A disadvantage in using a tungsten retorting crucible would be that the residual uranium product remaining after vaporization of the zinc and magnesium sometimes adheres to the crucible surface. Although operations at maximum retorting temperatures (800-900°C) do not result in attack of the tungsten, consolidation of the uranium product by melting (1150°C) in a tungsten crucible results in attack of the tungsten by the molten uranium. One possible means of freeing uranium from a tungsten crucible surface is by converting the uranium (at least in part) to the hydride. Also, by hydriding the product from the uranium-zinc intermetallic decomposition step (which precedes uranium dissolution), there is the possibility of eliminating the uranium dissolution step of the skull reclamation process.

altogether. It is possible that the partly hydrided uranium rather than uranium metal can be the final product of skull processing, and can be used as make-up material in the melt refining furnace. Experiments have been performed in tungsten crucibles to investigate these possibilities.

In one experiment, it was presumed that distillation of zinc and magnesium had already been accomplished and that a uranium product was adhering to the crucibles. In pilot-scale work, this has frequently, but not always, happened. To simulate this condition, the charge contained only natural uranium and fissium alloy. To insure adherence, the charge was melted at 1200°C. When cooled, the uranium metal was indeed stuck to the crucible. The hydriding step was then performed, and the entire product was easily dumped from the crucible. A silvery metal foil containing tungsten (~1 w/o of product) was not hydrided. The remainder of the product consisted mainly of beta-UH₃ in the form of a sintered cake (~5 w/o) and flakes (~94 w/o).

Another experiment was carried out to demonstrate hydriding of the product from the intermetallic decomposition step of the skull reclamation process. The charge was prepared by first simulating the zinc-uranium intermetallic formation and precipitation step of the process, then decomposing the intermetallic compound and subsequently precipitating metallic uranium. The product was metallic uranium dispersed in a zinc-50 w/o magnesium solution. This product was the charge for the third hydriding experiment. The maximum temperature of this hydriding run was 320°C. The hydrided product (fine powder and some larger metallic pieces) was dumped easily and was of low overall density (~2 g/cc). The two types of particles have been submitted for analysis. The potentialities of a hydriding step in the final product consolidation for the skull reclamation process will be further evaluated when the results of these analyses are received.

F. FARET

1. General

Principal activities for the current reporting period continued to be concerned with general Title II engineering at the Laboratory and at the architect-engineer's offices. Bechtel, whose efforts remain near peak level, report 65 percent of Title II complete at the end of August. The Laboratory's effort required for review of Bechtel's drawings and submittals has increased significantly.

Design Package II (reactor vessel) has been received by the Laboratory. Preparation of the ANL portion of the specifications and drawings has been completed, and the entire package consisting of the combined ANL-Bechtel input is ready for transmittal to pre-qualified vendors.

Design Package III (heat exchangers) and Design Package IV (sodium pumps) have been received and are being reviewed by the Laboratory.

A revised control schedule has been received by the Laboratory. Bechtel's current analysis indicates that there is no known reason why the remaining major design packages will not be submitted essentially on schedule.

Major submittals of drawings recently received include electrical schematics, sodium piping drawings, piping and instrumentation diagrams, etc. Recent transmittals to Bechtel by the Laboratory, in addition to routine comments and reviews, have included personnel air lock seal criteria, recommendations for rearranging the major argon system coolers so that they will not be in the high-level radiation environment, fuel-storage tank-internals criteria, shielding data, and miscellaneous comments on various ANL FARET Data Transmittal items. The ANL Data Transmittal items still outstanding are relatively minor, comprising only three of seventy submitted; delays because of these items are not expected.

Early procurement and construction plans of United Engineers and Constructors, Inc. (UEC) have been reviewed and refined as a result of joint effort on the part of the Laboratory and UEC. This work is continuing and currently is directed toward minimizing the impact of a single issue of Package IX on the UEC organizational effort requirements.

2. Safety Analysis

The revised FARET Preliminary Safety Analysis Report, ANL-6813 (Approval Copy), has been submitted to the Commission.

3. Reactor Vessel

The Reactor Vessel Selection Board has received and processed vendor responses to its request for a statement of interest and a submittal of qualifications for the work. The reactor vessel procurement package has been completed. Release of the package for proposal purposes is imminent.

4. Stress in FARET Support Grid Plate

The deflection and strength properties of a thin circular plate with two distinct concentric regions has been investigated mathematically. The deflection, radial, and tangential stresses were calculated on the assumption that the homogeneous plate deforms in accordance with Hirchoff's approximation.

5. Core Subassemblies

Design parameters covering the basic dimensional information have been prepared for the test zone and driver zone of the zoned core assembly (see Table VIII). These basic data will be used for further physics and thermal analysis. Similar work will be developed for the buffer zone.

Table VIII. FARET Fuel Data

	Test Zone (Slip Fit)	Driver Zone
Fuel Outside Dimensions	0.523	1525
Fuel Length	36	36
Cladding, Outside Dimension	0.593	0.221
Cladding Wall Thickness	0.033	0.017
Gas Gap Annulus Thickness	0.002	-
Elements per Subassembly	7	61
Subassemblies per Core	37	48
Sodium Bond Annulus Thickness	-	0.017
Spacer Wire Diameter	-	0.050
Spacer Wire Pitch	-	6
Fuel Area, in. ²	1.504	1.122
Fuel Area, Area %	32.27	24.07
Gas Area, in. ²	0.0231	-
Gas Area, Area %	0.496	-
Sodium Bond Area, in. ²	-	0.554
Sodium Bond Area, Area %	-	11.88
Sodium Flow Area, in. ²	2.370 (2)	1.889
Sodium Flow Area, Area %	50.83 (2)	40.53
Total Sodium Area, in. ²	2.370 (2)	2.442
Total Sodium Area, Area %	50.83 (2)	52.39
Cladding Area, in. ²	0.406	0.665
Cladding Area, Area %	8.71	14.267
Hex Can Area, in. ²	0.3118	0.3118
Hex Can Area, Area %	6.69	6.69
Spacer Wire Area, in. ²	-	0.1197
Spacer Wire Area, Area %	-	2.568
Total Metal Area, in. ²	0.765 (2)	1.096
Total Metal Area, Area %	16.4 (2)	23.5
Total Hex Cell Area, in. ²	4.661	4.661
Number of Spacers	5	-
Material	Pu ²³⁹ -U ²³⁸ O ₂	Enriched Uranium Fissionium
Enrichment a/o	12.5 (Pu)	80 (U ²³⁵) Est.
U ²³⁵ , kg	-	435
U ²³⁸ , kg	276	108
Pu ²³⁹ , kg	39	-

NOTE: 1. All dimensions in in. except where otherwise noted
2. Weighted to include spacer volumes

6. Instrument Electrical Connectors

Electrical leads, male and female terminals, and insulating components are commercially available for the instrument connections with the fuel elements. A configuration and preliminary prototype of a complete connector assembly had been developed (see Progress Report for July 1963, ANL-6764, p. 29). This prototype consists of a plug attached to a bundle of leads which are enclosed in a metal hose and a receptacle which is part of the fuel assembly. It should be possible to assemble the plug and receptacle by means of remote manipulation.

It has been possible to develop the mechanisms to execute the desired remote connection by means of the fuel-handling machine. This mechanism includes a special grip and locking tool to be carried and operated by the fuel-handling machine. A preliminary prototype assembly has been constructed and tested.

The solid sealing lip of the preliminary prototype connector required a tightening torque of 40 ft-lb. The seal could not be made consistently tight at this clamping force. A more compliant (less rigid) seal will be incorporated in the connector (see Figure 20).

Figure 20. Sealed Connector

A tool to latch on to the body, Item 7 in Figure 20 (at the groove near the joint with the hose, Item 6) is shown in Figure 21. This tool rotates the locking sleeve, Item 3 in Figure 20, clamping the connector plug firmly to the fuel element, Item 1; at the same time rotation of the tool operating jaw disengages the latch releasing the installed connector from the grip of the tool.

Figure 21. Connector Handling Tool

Finally, a tentative design for the connection of the external cable with the in-core leads at the extension piece flange is shown in Figure 22. This assembly is designed to be replaced with relative ease as when leads inside the vessel become damaged or have to be replaced for other reasons.

Figure 22. Connection of External Cable with In-core Leads

Components shown in the figures are being fabricated for testing and evaluation. Tests of several types of commercial gaskets are in progress to decide on the most promising selection.

7. Fuel Assembly Sodium Flow Test Facility

Detailed drawings of the buffer and driver and control subassemblies for the flow test facility have been completed. Design work continues on the test zone elements required. This work entails the provision for instruments and connectors necessary for the in-core instrumentation. All assemblies except for the test zone elements have been ordered.

The status of the construction of the test loop is as follows:

a. The rework of the three vessels for the Flow Test Loop is progressing satisfactorily. The dump tank and expansion tanks are to be shipped the end of September. Shipment of the pressure vessel is scheduled for the end of October. It was necessary to compensate dimensionally on the parts that will fit into the reactor vessel due to dimensional errors made on the vessel during partial fabrication, so that the repairs could be made expeditiously.

b. Twenty pipe fittings were rejected upon delivery from the vendor because they were made of 304L instead of 304 stainless steel. This replacement will cause a serious delay in the piping fabrication.

c. Changes have been added to the Byron Jackson pump order.

d. Radiographs of the interim pump case disclosed several welding areas which must be repaired. It is not known if these areas are caused by previous operation or if these were original faults.

8. Sodium Deposition Test

A study has been made to evaluate the amount of sodium vapor that would condense on the cooler sections of the FARET reactor vessel. Within the vessel the ambient temperature will be as great as 1200°F; external to the vessel the ambient temperature is about 100°F. At 1200°F, the sodium vapor pressure is greater by several orders of magnitude than in sodium-cooled reactors operating at temperatures of 700-1000°F. Accordingly, sodium deposition on the cooler surfaces is expected to increase. Correspondingly, problems associated with sodium deposition on bellows; small crevices, clearances, bearing and seals; vent and purge lines, instrumentation conduit; etc., are expected to be aggravated.

In order to determine the magnitude of sodium deposition, a facility comprised of a sealed vessel, 12 in. in diameter, approximately 36 in. in height with a 4-in.-diameter standpipe extending vertically.

upward for 7 ft, was constructed. A 2-in.-diameter pipe is located concentrically within the 4-in.-diameter vertical section and extends downward into the 12-in.-diameter section. Eleven horizontal surfaces (discs) are welded to the upper end of this 2-in. pipe at approximately equal intervals over an axial distance of 30 in., so as to simulate crudely a FARET control rod drive shaft. Included in the 4-in.-diameter section are three surface sample holders placed on a common vertical line 18, 37, and 56 in. above the sodium level. These sample holders contain 3/4-in.-diameter surfaces machined at different angles to the vertical to create diagonal elliptical surfaces. Sample surfaces sloped at 30°, 45°, and 60° to the vertical were tested.

The vessel was filled with sodium to a predetermined level, and the temperature raised and held between 1100 and 1200°F. The temperature of the 2-in.-diameter mock control rod drive varied from 1200°F at the sodium level to room temperature at the top of the standpipe. The average temperature of the three surface samples were 390, 245, and 190°F from bottom to top, respectively.

The facility was operated for approximately 7,000 hr. During this time, six inspections were made. The sequence of the inspections were as follows: After operation for a predetermined length of time at a near-constant sodium temperature of 1200°F, the area samples were removed without disturbing the mock control rod drive shaft. The amount of sodium deposited on a sample was determined by a titration technique, and the samples were cleaned and returned to their holders. Operation was then continued at approximately the same temperature for an equal length of time. At the end of this period, both the surface samples and the mock control rod were removed, and a visual inspection of the rod and the standpipe was made. Table IX lists results.

Significant quantities of sodium deposited in the standpipe of the vessel in Run 2 (see Figures 23 and 24). Large solid deposits of sodium were found at the 34 to 40 in. level and on the horizontal surfaces of the shaft. The two higher (and cooler) surface samples (see Figure 25) were completely covered with sodium; the bottom had large beads of sodium on the surface. No attempt was made to evaluate the amount of sodium because of the excessive quantity present. The second rod removal inspection (Run 4) showed nearly identical results.

After Run 4, a thermal barrier, 4 in. long with a 0.020-in. radial clearance, was placed into the standpipe to reduce the area available to thermal gas flow. Run 5 showed results similar to those of Run 1. When the rod was removed after Run 6, the condition was similar to that found in the other two rod inspections. Analyses of the 60 and 45° sample were made, but the 30° sample was covered with excessive sodium. Because of the variation in the data, it cannot be concluded that the thermal barrier caused any noticeable reduction in the amount of material transferred.

Table IX

Run No.	Duration of Operation, hr	Sodium Temp, °F	Sample Information				
			Type of Sample	Distance above Sodium, in.	Sample Temp, °F	Rate of Buildup, in./y	Type of Inspection
1	1200	1200	60°	18	388	0.01	Removed Area Samples Only
			45°	37	244	0.0024	
			30°	56	189	0.00125	
2	1200	1200	No Analysis Made				Removed Area Samples and Rod
3	790	1100*	60°	18	389	0.10	Removed Area Samples Only
			45°	37	245	No Analysis Made	
			30°	56	189	0.0025	
4	790	1200	60°	18	437	0.23	Removed Area Samples and Rod
			45°	37	395	0.83	
			30°	56	158	0.378	
5	1512	1130*,**	60°	18	434	0.0086	Removed Area Samples and Rod
			45°	37	352	0.0047	
			30°	56	140	0.0006	
6	1368	1200**	60°	18	436	0.007	Removed Area Samples and Rod
			45°	37	369	0.0016	
			30°	56	152	No Analysis Made	

* Loss of Heater.

** Thermal Barrier Installed.

Figure 23. Sodium Deposition in the Mock Bellows Area of Shaft

Figure 24. Sodium Deposition at the 34- to 40-in. Level on Shaft

Figure 25. Surface Samples Viewing, from Left to Right,
56 in. - 30° , 37 in. - 45° , 18 in. - 60°

III. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. In-core Fast-neutron Spectroscopy

Recoil-proton distributions were taken with proportional counter probes (see Progress Report for July 1964, ANL-6923, p. 47), of 2.5-cm diameter, 6.9-cm effective length, and 0.12-cm thick stainless steel walls, placed near the center of the ZPR-VI core. The probes were centered in the 5.1-cm by 5.1-cm ZPR-VI drawer by a lead block which just filled that part of the drawer containing the probe itself. Insertion of the probe into the core necessitated the removal of one complete fuel drawer. However, the probe drawer could have been loaded with core material to about half of its total volume although this was not done.

Measurements were made in Assembly No. 3 of ZPR-VI. The flux was held at a fixed level by operating at about 2% subcritical and with startup sources in place. This condition produced a detector count rate of less than 1500/sec. With a multiplication of about 50, the spectrum should be very close to that for the assembly at critical.

The final critical configuration (shape: 51-cm long cylinder of 77-cm radius) is described in Table X.

Table X. Specifications for Assembly No. 3 of ZPR-VI

	Core Atom Density ($\times 10^{24} \text{ cm}^{-3}$)	Core Volume Fraction	Blanket Atom Density ($\times 10^{24} \text{ cm}^{-3}$)	Blanket Volume Fraction
U ²³⁵	0.002285	0.0476	8.104×10^{-5}	0.001688
U ²³⁸	0.009853	0.2052	0.03993	0.8318
SS-304	0.01475	0.1751	0.006003	0.07122
C	0.01290	0.1540	-	-
Na	0.00791	0.3595	-	-

The energy region below 100 keV was measured by a counter filled to 3 atm of H₂, about 1/3 atm of CH₄ for quenching, and an equal pressure of N₂ for calibration. Above 100 keV, another counter having identical dimensions but filled to 3 atm of CH₄ and 10 cm of N₂ was used in order to reduce end and wall-effect distortion. The range of a 0.75-MeV proton in the methane counter is about 0.5 cm. One expects a significant correction in the spectrum would be necessary at this energy and above, but no corrections were made here. Such a correction would diminish rapidly as energy decreased, however, and probably would not have been very serious below about 500 keV.

Data were taken with a two-parameter pulse-height analyzer, one parameter for ionization and the other for rate of pulse rise. Each parameter was digitally converted to 32 channels full-scale. Examination of the distribution of the rate of rise of pulses permits events of a given ionization to be sorted into two kinds, one due to gamma rays and the other due to neutrons. In this way, a recoil-proton ionization spectrum was derived free from gamma contamination. Because of the finite number of channels in the analyzer, signal/noise ratio, and other reasons, the interesting energy region (1 keV to 1 MeV) was observed in sections, each changing by a factor of about 3 in energy. The energy scale corresponding to different counter voltages had already been calibrated against the $N^{14}(n,p)C^{14}$ reaction.

The neutron spectrum derived by differentiation is shown in Figure 26. (The relative neutron flux per unit lethargy is $E^2/\sigma(E)$ times the derivative.) Included in Figure 26 for comparison are the results of a group calculation for this assembly. The experimental spectrum is "softer" than the group calculation.

Figure 26. Neutron Flux per Unit Lethargy vs. Neutron Energy. The resolution of the measurement, which depends upon energy, is indicated by the length of the base of the triangle. The resolution is shown at three different energies. The vertical bars through some of the experimental points show the range of the experimental error. The area under the experimental curve was normalized to equal the area under the histogram.

2. High-conversion Critical Experiment

The feasibility of a new technique for the determination of the effective capture-to-fission ratio in undermoderated low-flux reactor assemblies was demonstrated in one of the systems studied in the High Conversion Critical Assembly Program. Measurements of the relative amplitudes of reactor response to oscillated samples of Cf^{252} and Mn, combined with a separate determination of the spontaneous fission neutron emission rate of the Cf^{252} and the absolute absorption rate of Mn, yield the relative importance of fission and reactor neutrons. This information, coupled with the measured effect of an oscillated U^{235} sample and the number of neutrons per U^{235} fission, yields a value for $\bar{\sigma}_c/\bar{\sigma}_f$ for the reactor neutron spectrum.

A provisional value of 0.25 has been obtained from the first complete set of data obtained in an assembly of Hi-C aluminum-clad fuel pins having a H:U atom ratio of 2.93. This value compares favorably with a semi-empirical value of 0.246 obtained from the U^{235} fission cadmium ratio measured in this core, the accepted value for σ_c/σ_f in a thermal flux, and a calculated value for the epithermal σ_c/σ_f .

This preliminary value obtained using the pile oscillator technique as well as its estimated error, will be influenced by a number of factors, some of which are now being investigated. The uncertainty in the strength of the spontaneous fission source will be reduced by a more accurate determination of the influence of hydrogen captures in the manganese-bath calibration procedure. The reactivity effect of the oscillated samples has been approximated by using only the first-order terms in a Fourier series fit to the experimental data. Any excess signal from the constant Cf^{252} source, during its insertion into and removal from the reactor, compared with the reactor response to a sample whose signal depends on the flux along its path from above the top reflector to the core center, has not, as yet, been deducted. Corrections are also required for other effects which are thought to be less significant. These include the difference in average fission neutron energy for Cf^{252} and U^{235} , reactivity effects of impurities in the manganese sample and other isotopes in the enriched uranium sample, differences in the neutron energy-dependent absorption of manganese and U^{235} , difference in the effect produced by point and distributed samples, and perturbation of the core spectrum by the tube in which the samples are oscillated.

B. Theoretical Reactor Physics

1. Resonance Interference

The exact solution of the stationary, infinite medium slowing-down equation involves an integral over a different lethargy range for each isotope in the medium. This raises difficulty in solving the equation by forward integration since either the number of values kept in the memory must be high or else the amount of tape handling must be increased.

The Laplace transform can be used to alleviate this problem. The resulting equation is

$$\tilde{F}(p) = \sum_{i=1}^N \left\{ \left[\int_0^p \tilde{A}_i(p-p') \tilde{F}(p') dp' \right] \left[1 - e^{-p\Delta_i} \right] + \frac{\sigma_{sio}}{(1-\alpha_i)E_0} \left[\frac{e^{-p\Delta_i}}{p(p-1)} - \frac{e^{-\Delta_i}}{p-1} + \frac{1}{p} \right] \right\} \quad (1)$$

Terms in Eq. (1) with the subscript i are isotope dependent, and the tilda implies the Laplace transform variable. The quantity $F(u) = L^{-1} [\tilde{F}(p)]$ is the collision density. Equation (1) can be solved by direct forward integration provided $\tilde{A}_i$ is known. Finally, by taking the inverse transform, $F(u)$ can be found.

A systematic test was made using numerical programs to find the Laplace transform and its inverse. These tests were based on the work done by Salzer⁵ and Shirliff and Stephenson.⁶ The tests were designed to check the accuracy of the method when successive transforms were made of standard test functions and a special function chosen to represent a typical resonance term. The method involves the use of polynomial fits to the given function.

The tests revealed these points:

- a. Round-off error is considerable, and the retention of 15 significant digits is necessary to secure 4-place accuracy in the transform of a 10th-order polynomial.
- b. To minimize error, the inverse transforms must involve polynomials which are one order higher than the order used to take the transform. This is a consequence of the design of the method. Moreover, as Shirliff and Stephenson show, error in taking the inverse is a strong function of the order of the polynomial. This may make it difficult to control the error in the final result.

By use of double-precision arithmetic for both the transform and its inverse, and use of 6- and 7-point formulae, agreement between the results obtained from special test function and the inverse transform of the transform could be retained to six or seven places over the lethargy range (0,1).

It is therefore concluded that the basic method is feasible but that more careful control of error propagation and stability is required.

⁵H. E. Salzer, Tables for the Numerical Calculation of Inverse Laplace Transforms, J. Math. Phys., 37 (1958).

⁶C. J. Shirliff and D. G. Stephenson, A Computer Oriented Adaption of Salzer's Method for Inverting Laplace Transforms, J. Math. Phys., 40 (1961).

2. An Altered Equation of State for AX-1

The calculation of energy yield by the AX-1 code⁷ is usually performed with the simple equation of state proposed first by Stratton.⁸ The equation has a number of parameters which by proper adjustment approximates the actual behavior of reactor material for small changes of density. The energy yield obtained with AX-1 is probably accurate for large excursions in fast reactors. At the present time the emphasis is shifting toward the prediction of energy yield for smaller and more probable excursions, along with a prediction of the damage suffered by a reactor in such an excursion. In terms of this point of view, Stratton's equation has been re-examined and some modifications have been proposed.

Stratton's equation assumes that pressure p is a linear function of density ρ and temperature θ :

$$p = \tau + \beta\theta + \alpha\rho \quad \text{for } p > 0. \quad (1)$$

Pressure is assumed to vanish when the above expression gives negative values. The specific heat at constant volume, c_v , is also assumed to be a linear function of temperature:

$$c_v = A + B\theta, \quad (2)$$

for both positive and vanishing pressures. The latter assumption is logically inconsistent. From thermodynamics we know that the changes of internal energy e with volume v is given by

$$\left(\frac{\partial e}{\partial v}\right)_\theta = \theta \frac{\partial p}{\partial \theta} - p.$$

Thus

$$\frac{\partial c_v}{\partial v} = \frac{\partial^2 e}{\partial \theta \partial v} = \theta \frac{\partial^2 p}{\partial \theta^2} \quad (3)$$

As there is an abrupt change in $\partial p / \partial \theta$ going from zero pressure to positive pressure, there must be also an abrupt change in the specific heat. When c_v is given by Eq. (2) for positive pressures, straightforward calculation gives

⁷D. Okrent et al., AX-1, A Computing Program for Coupled Neutronics-Hydrodynamics Calculations on the IBM-704, ANL-5977 (1959).

⁸W. R. Stratton, T. H. Colvin, and R. B. Lazarus, Analysis of Prompt Excursions in Simple Systems and Idealized Fast Reactors, Proc. 2nd Geneva Conf. 12, p. 196 (1958).

$$c_v = A + B\theta + \frac{\alpha\beta^2\theta}{(\tau + \beta\theta)^2} \quad (2a)$$

for vanishing pressures. Although this modification produces only a small difference in calculations during heat generation, it becomes important when voids are formed following a sharp burst. The neglect of this modification produces an inconsistency in the determination of energy and results in an unduly high code-control parameter ("error local"). This tends to slow down numerical calculations and probably causes sharp instabilities in the calculated pressure wave.

This simple modification is not sufficient to make the calculation of the pressure wave realistic. Shock experiments show⁹ that for sufficiently large pressures e of uranium is a linear function of pressure. Thus the equation of state can be written essentially in "Bethe-Tait" form:

$$e = Q^*(v) + \frac{v}{(\gamma-1)} p, \quad (4)$$

where Q^* , the threshold energy density, is a function of specific volume v . It increases with v approximately to the heat of vaporization ($L \approx 2 \times 10^{10}$ erg/g). The parameter $(\gamma-1)$ is also a function of v , continuously decreasing down to about $1/3$. Stratton's equations, after elimination of temperature, can be written in essentially the same form.¹⁰ However, $(\gamma-1)$ increases rapidly with increasing v , and Q^* increases to infinity with increasing v . Thus a change of Stratton's equations of state, when large changes of density (e.g., cavitation) are expected, seems mandatory.

The equation of state (4) is still not valid when liquid uranium is in equilibrium with uranium vapor. The proper equation of state in this case is

$$e = v \left(\theta \frac{dp}{d\theta} - p \right) + A'(\theta), \quad (5)$$

where θ is determined by saturated vapor pressure. The temperature is approximately given by¹¹

$$\theta = (L/R) \ln(p_1/p), \quad (6)$$

where L is the heat of vaporization, R is the gas constant, and the pressure p_1 is of the order of half a megabar. Physical considerations lead to the equation of state, Eq. (4) or Eq. (5), whichever gives a larger value for e .

⁹E. Morris, The Equation of State of Uranium in the Critical Region, AWREO-61/62.

¹⁰V. Z. Jankus, A Theoretical Study of Destructive Nuclear Bursts in Fast Power Reactors, ANL-6512 (1962).

¹¹E. G. Rau and R. J. Thorn, Vapor Pressure of Uranium, ANL-5203 (1954).

In a large reactor, the Doppler effect is expected to contribute significantly to the shutdown. The Doppler effect is primarily determined by the temperature of the material. The latter is determined in the saturated vapor region by Eq. (6) and can be obtained in "Bethe-Tait" region, Eq. (4), by progressing along each adiabat. Applying the second law of thermodynamics to Eq. (4) above, we obtain

$$\theta \exp \int (\gamma-1) \frac{dv}{v} = F \left[(e-Q^*) \exp \int (\gamma-1) \frac{dv}{v} + \int Q^{*1} dv \exp \int (\gamma-1) \frac{dv}{v} \right], \quad (7)$$

where F is an undetermined function. It can be evaluated, however, by applying Eq. (7) along the intersection of saturated vapor and "Bethe-Tait" regions.

In an actual reactor with a reasonable amount of voids, the "Bethe-Tait" region is reached at fairly high temperatures; the Doppler effect at these temperatures is expected to be small. Thus it seems sufficient to approximate F in Eq. (7) by a linear function (assume that specific heat is constant). Then F can be obtained quite simply in the numerical program whenever the given material initially reaches the "Bethe-Tait" region. The temperature determined in this way is not expected to be completely correct in the subsequent wave propagation (when the densities have changed substantially). However, at this time, reactor power is expected to be negligible, and determination of the Doppler effect and reactivity is of no consequence to the subsequent purely hydrodynamic calculations.

C. High-temperature Materials Development

1. Ceramics

a. The Preparation of Uranium Monosulfide. An effort to synthesize uranium monosulfide electrolytically is being made. In initial attempts involving the electrolysis of UF_4 and Na_2S in a fused salt bath composed of $LiCl$ and KCl at $525^\circ C$, the resulting product was a mixture of alkali metal, U , UO_2 , and UOS . Experiments were therefore undertaken to establish the sources of contamination and to modify the experimental procedure so as to obtain the desired product.

The salt-bath composition was changed to $NaCl-KCl$ in order to permit operation at higher temperatures and to avoid the extreme hygroscopicity of $LiCl-KCl$ mixtures. In addition, the apparatus was modified to allow stirring of the melt in order to reduce polarization and prevent alkali-metal formation at the cathode. Preliminary experiments at $700^\circ C$ indicated that this measure was effective.

The main experimental problem appears to be contamination by water-vapor, which is probably responsible for the high-oxygen contamination of the product as well as for undesirable side effects, such as excessive

evaporation of the salts. Attempts will be continued to achieve dryer conditions and to optimize operating parameters such as temperature, reaction time, and applied emf.

b. Properties of (Th,U) Phosphides. The results of research this month show that ThP and UP do not exhibit complete mutual solubility across the entire compositional range. In addition, it was found that the intermediate compositions in the ThP-UP system exhibit slightly greater weight losses in vacuum at 2200°C than do the end members.

Three compositions in the ThP-UP system were prepared and fired at 1800°C in vacuum. Little interaction occurred at this temperature and the samples were later fired at 2000°C until maximum interaction occurred. X-ray analysis showed single phases at both 75% ThP-25% UP and 25% ThP-75% UP, and two phases at the 50% ThP-50% UP composition. Thorium phosphide appeared to take up about 40% of UP, whereas UP only took up about 25% of ThP. The diffuse patterns indicated considerable lattice strain, particularly in the UP-rich end of the system. The absence of complete solubility between ThP and UP is puzzling in view of the fact that ThS and US exhibit complete mutual solubility. The two lattice constants obtained for single-phase material showed a positive deviation from a linear relationship. The same phenomenon was observed in the case of UP-US and ThS-US solid solutions.

The three aforementioned compositions and the two end members were fired in vacuum at 2200°C to obtain weight-loss data at elevated temperature. The results are given in Table XI, showing that the intermediate compositions exhibited greater weight losses than the end members, and that ThP was more volatile at 2200°C than UP. Greater weight losses were also noted for the intermediate compositions in the UP-US system.

Table XI. Thermal Weight Losses of (Th,U)P Phosphides

Composition:	ThP	75% ThP- 25% UP	50% ThP- 50% UP	25% ThP- 75% UP	UP
Weight Loss/hr, %	2.06	2.54	3.26	2.2	1.24

The sintered densities of the intermediate compositions were generally lower than those of pure end members.

c. Mechanical Properties of Ceramic Uranium Compounds. Checking an alignment of the high-temperature furnace attached to the Riehle machine revealed a leak in the furnace shell, and the whole equipment has been returned to the makers for repair. The tungsten bending tools were tested with copper specimens, and proved successful.

A start has been made with investigation of the processing variables of UO_2 , although testing at room temperature provides the only means

of assessment. As a result of these tests on pressing conditions and sintering temperatures, 96% theoretical density in specimens breaking with a modulus of rupture of over 11,500 psi has been achieved to date.

d. Anelasticity of Some Group V and VI Uranium Compounds

(i) Apparatus for Dynamic Measurement of Elastic Modulus and Internal Friction. In the last report (Progress Report for August 1964, ANL-6936, p. 53) it was mentioned that the internal friction results obtained for a sapphire rod were higher than the one found in the literature.¹² That was attributed to air damping and to the position of the suspension wires. In order to verify that, measurements were preformed by placing the wires at different positions. When the wires were far from the nodes (31 mm), the damping was very high; a minimum was obtained when placed at one millimeter from the node. This result is in fair agreement with the one given by Wachtman and Lam.

A circuit was incorporated to the apparatus. It shuts off the driving signal and simultaneously triggers the sweep of the oscilloscope.

(ii) Preparation of Samples. The O/U ratio of the hot-pressed rods was 2.05. A similar test made in the original powder gave a value of 2.14. The microstructure of the hot-pressed samples was different from that of one of the pellets sintered in a hydrogen atmosphere. The amount of porosity is larger and the grain size seems to be very small.

Four bars of UO_2 of rectangular cross section have been pressed to a green density of about 50% of theoretical density and sintered in hydrogen at 1750°C for 4 hr. They will be sent for machining.

(iii) Control of Sample Parameters. Pellets have been pressed and are ready for sintering. They will be used to check some of the results obtained previously.

2. Corrosion by Liquid Metals

a. Lithium Corrosion at Elevated Temperatures. The corrosion study of refractory metal-lithium systems at high temperatures has continued, since the low density, high specific heat, and high boiling point of lithium are properties likely to recommend it for use as a coolant.

Tantalum, one of the potential containment materials for high-temperature liquid metals, has good high-temperature strength and

¹²Wachtman, J. B., Jr., and Lam, D. G., Jr., Young's Modulus of Various Refractory Materials as a Function of Temperature, J. Am. Ceram. Soc., 42 (5), 254-60 (1959).

workability. However, very little information is available regarding its resistance to lithium above 1000°C.

Two recent capsule tests of powder-metallurgy-product tantalum with lithium at 1000°C for 168 hr have indicated that this material is susceptible to transgranular attack. The attack on the inner walls of the capsules adjacent to the bottom was rather severe. There was no visible corrosion above the liquid level. A discontinuous oxide layer was noted on the outer surface of the capsule, which suggests that a 5×10^{-6} torr vacuum environment at the temperature was not sufficient to protect the material from oxidation. Use of a getter material for future tests is under consideration.

The powder-metallurgy-product tantalum has a 99.9+% purity and its density is of the order of 97% theoretical. The impurity and porosity of this material may contribute to its low resistance. High-purity, arc-melted material is now available. Further tests with the latter material will test the above speculations.

D. Other Reactor Fuels and Materials Development

1. Nondestructive Testing

a. Determination of Elastic Moduli of High-temperature Materials by Ultrasonics. The object of this program is to measure the temperature dependence of the elastic moduli of materials that show promise in high-temperature reactor applications. To obtain these data a vacuum furnace has been constructed in which solid bar-shaped samples will be mounted. Pulsed ultrasonic waves will be propagated along the length of these samples, and the transit times of pulses passing a heated, indexed region in each bar will be measured. The indexing is done by machining one end to a smaller diameter than the rest of the bar.

The velocity of sound in the indexed region can then be calculated from a knowledge of the length of this region and the transit time in it. Both shear wave velocities and longitudinal velocities are determined.

By a method whereby measurements of two parameters can be made at the same time (see Progress Report for April 1964, ANL-6885, p. 45), both the longitudinal and shear wave velocities may be calculated from one run. This scheme made use of rods having sharp threads cut into them in order to disrupt the pattern of trailing pulses produced by the ultrasonic wave-guide effect. Reproduction of work done by Dunegan¹³ has shown that, although the trailing pulses are not entirely eliminated by the

¹³Dunegan, H. L., Determination of Dynamic Modulus at High Temperatures by the Use of Ultrasonics, UCRL-7326 (Rev. 1) (July 18, 1963).

threading technique, they can be reduced, by proper design of the specimen, to such small amounts that the desired echoes from the specimen are not obscured. This technique will be adopted.

Measurements of longitudinal wave velocity and of delay time of trailing pulses were made in a bar of vanadium-20 w/o titanium alloy at room temperature to determine the dimensions of the bar in its final form as a specimen to be placed in the vacuum furnace.

In order to test the ultrasonic and high-temperature systems, measurements of sound transit times were made from room temperature up to 930°C in a stainless steel specimen at 5 Mcps. This specimen was one in use as a "dummy" for many months, and its surface condition was far from ideal. Furthermore, its surface was not threaded, so that it was necessary to make two temperature runs in order to measure both longitudinal-wave transit times and shear-wave transit times. From the measured transit times plus known values of thermal expansion, the longitudinal-wave velocity and the shear-wave velocity were calculated as a function of temperature. From these, plus a knowledge of the density, the elastic moduli can be computed.

Since the specimen was not in the best of condition, the sound pulses were not very well-shaped, and uncertainties in the data are due to a large extent, to this factor.

Another possible source of errors is a thermal gradient in the sample. In the particular run described above, the largest temperature difference between two thermocouples placed 2.2 cm apart was 16.6°C at 530°C. It was found that the spot-welded thermocouple junctions gave false readings, probably because of poor heat conduction through the rather porous bead region. Better consistency in thermocouple readings has been obtained since the junction and the attachment to the specimen have both been made at the same time by resistance welding under pressure. However, the thermal gradient is still definitely present in the specimen. Attempts at reducing this gradient have failed so far because the thermal emission was such that too little heat was transferred to the sample. A coil wound directly on a mica-insulated sample shows more promise and is being studied further. The vanadium-20 w/o titanium sample, and subsequent samples, will be measured in spite of any gradient present.

Room-temperature measurements of longitudinal wave velocity and of delay time have been made on a tungsten bar preparatory to its being made into a specimen.

b. Ultrasonic Imaging. Defect areas located in several test samples are now being subjected to metallographic tests. Samples include aluminum-clad aluminum-uranium alloy, rolled ALPR stock, and several

of the prototype A^2R^2 fuel plates. Additional ultrasonic tests on some of these latter samples are planned after receipt of some useful 1-Mc transducers for use with the imaging system.

c. Development of a Neutron-image-intensification System. Flat neutron-scintillator screens of various thicknesses, made by methods similar to those expected to be used to prepare the scintillator input screen in a neutron-image-intensifier tube, have been tested to determine the light emission for a given neutron exposure and to determine the neutron-attenuation properties. The peak light emission was obtained for total screen thicknesses between 0.4 and 0.6 mm, which transmitted approximately 70% of the incident neutron intensity when use was made of the Juggernaut reactor beam (cadmium ratio, 2.4).

These preliminary tests indicate that the input screen for the intensifier tube should have a thickness of about 0.5 mm for optimum light emission. Preparations to construct a complete neutron-image-intensifier tube are now being made. Equipment to operate such a tube when it is received has been collected at the Juggernaut reactor. This includes the power supplies to operate the intensifier tube and a closed-circuit television system to bring the neutron-image information out of the radiation area.

d. Thermal Conductivity of Irradiated Fuel. Apparatus is being assembled to measure the thermal conductivity of irradiated metallic fuel specimens as a function of burnup so that fuel center temperatures during irradiation may be calculated more accurately. Measurements will be made on segments of the metallic pins customarily used as irradiation specimens.

Two measurement systems can be applied. In one, use is made of a modified Kohlrausch method; it has been tested on various homogeneous specimens and is capable of giving consistent results within about 5% of published values for these materials. However, this method has the disadvantage of requiring specimens with a minimum length of 2.5 cm. If appreciable differences of conductivity occur along the irradiated pins as expected (because of temperature and flux variations), the measurements by this method will become quite complicated. The Kohlrausch method measures axial conductivity, which might be a serious disadvantage since there is no a priori assurance that the conductivity in these pins will be isotropic. For calculation of center temperatures a radial value of conductivity would be preferred.

In the other system, the flash method, the conductivity in plane samples is measured. There was some question as to whether accurate measurements could be made on specimens of the size obtainable from the irradiated pins, but trial measurements have shown that this is possible. With the flash method; the conductivities in both the axial and in the radial

directions can be measured at a considerable number of points along the irradiated specimen. This will permit a more accurate plot of thermal conductivity as a function of distance along the pin. The main disadvantage seems to be the number of small flat specimens required, which for a number of reasons will be difficult to fabricate remotely, and the careful experimental technique necessary, which does not particularly lend itself to cave operation.

e. Infrared Imaging. Some initial motion picture recordings from the infrared television system as various test objects were subjected to heating and cooling cycles have been made. The results were reasonably successful in that several test samples did produce maximum transient indications of internal defects during portions of the test. Further studies of this nature are planned.

E. Remote Control Engineering Development

1. Viewing Systems

The study of certain elements added to simple glasses in small concentrations is being carried out to understand the effects of radiation on glass and to develop radiation-resistant glasses. Recent studies have used high-purity fused silica as the base glass, whereas previous work used mainly alkali aluminoborate glasses.

Thallium is being studied because it forms both electron and hole traps in glass similar to cerium. Furthermore, there is a considerable understanding of its properties in crystals. Before irradiation, thallium forms two absorption bands in fused silica. The absorption band at $213\text{ m}\mu$ is attributed to Tl^+ and a much weaker band at $253\text{ m}\mu$ to aggregates of two or more Tl^+ .

Iron is of interest also because it forms both electron and hole traps and is a common impurity in silicate glasses. An absorption band in sodium silicate glass at $225\text{ m}\mu$ is attributed to Fe^{3+} . The corresponding band in aluminoborate glass is at $220\text{ m}\mu$.

Work is in progress to set up equations relating some of the parameters in the suppression of color-center formation in glass due to addition of an element like cerium. Also, equations relating the rate of variation of optical density, conductivity, and thermoluminescence with temperature are being developed.

2. Electrical Fracture in Glass

Previous destructive measurements on large samples of glass have shown that different compositions of glass have different susceptibility to

fracture by electrical discharge. As an aid in developing a discharge-resistant glass, and expedient test that can be performed on small samples is desired, which can be correlated with the destructive tests on large samples. Two types of tests, both of which essentially measure charge conductivity, are being carried out. One is the conventional measurement of volume resistivity at low voltage. The other is the measurement of the decay of charge injected into the glass by electron irradiation.

A new nondestructive method of performing this latter measurement may reduce the number of samples needed for each experimental glass composition. Preliminary results with both tests correlate qualitatively with the test on large samples. More work is in progress to develop a satisfactory test.

3. Electric Master-Slave Manipulator Mark E4

Work has continued on construction of some of the servo amplifiers. Four gearboxes have been completed with the exception of synchro drives, which are designed but not yet fabricated. Design work on the shoulder joint of the manipulator arm has continued on both an offset arrangement (that is, where the upper manipulator arm would be offset from the arm connecting it to the servo drive packages) and an inline arrangement.

Two different counterweighting systems have been studied for the inline arrangement. One of these uses two separate counterweights which incorporate a force inverter. This latter method is interesting in that it is more compact at the shoulder joint. It, in effect, applies a force always in the opposite direction to gravity.

4. Special Motors for Master-Slave Manipulators

The aluminum cup-rotor motor (see Progress Report for July 1964, ANL-6923, p. 62) has been tested further, and two unforeseen characteristics showed up. One is the intermittent rubbing of the rotor against the stator, and the other is a maximum speed less than that which was expected. The first can be corrected by increasing the clearance of the cup and improving alignment of some of the parts. The second is believed to be caused by the large spacing between the stator teeth. This large spacing was designed into the motor to provide cooling air channels and to keep the leakage reactants at a moderate value. At present, the inside stationary core is also toothed; it is believed that some improvement in free speed can be achieved by using a smooth core.

5. Higher-fidelity Manipulators

Studies are being carried out on methods of achieving higher fidelity in master-slave manipulators. A test device has been partially constructed

to introduce various magnitudes and rates of change of mechanical position to a handle. Tests with this should give some information on the detectability of the human operator.

Another device has been partially constructed which has a spring between the probe and the handle. Tests with this apparatus should give some information on the frequency range needed.

F. Heat Engineering

1. Two-phase Flow Studies

a. Void Fraction - Pressure-drop Facility. The loop of the pressure-drop facility has logged an additional 150 hr at 750°F and 450 hr at 1000°F with no indication of corrosion or excessive oxide impurity as indicated by the electromagnetic flowmeters. Several gamma-attenuation traverses have been taken across the adiabatic pressure-drop section, and show good stability and a reasonable empty-to-full ratio. As soon as the condenser heat-loss runs are complete, low-pressure boiling data will be obtained at approximately 1450°F to test the stability of the homogeneous boiler and the differential pressure measurement system.

2. Boiling Liquid Metal Facility

a. Niobium-1% Zirconium Loop. This facility is designed to investigate the heat transfer and two-phase flow characteristics of boiling sodium to a temperature of 2100°F, at which the pressure is approximately 8 atm. Among the variables to be investigated are boiling heat flux and temperature difference, boiling and adiabatic two-phase pressure drop, vapor volume fraction, and boiling stability of the proposed loop configuration.

The one-piece loop and pump tube structure design is complete, and the purchasing process has been initiated. The loop will be constructed of Nb-1% Zr alloy, mostly 1/2-in.-OD tubing, exclusive of the thermal radiation heater and the pump duct.

The helical induction pump that has been ordered will provide for flow rates up to 6.5 gpm at a 20-psi head. This will provide a superficial velocity of liquid sodium up to approximately 15 ft/sec in the high heat flux heater section. Dependent upon the ultimate heater capability, the entire quality range from 0 to 100 percent will be investigated at as high a flow rate as practicable. The thick-walled boiler will be heated initially by a tungsten radiation heater over a heated length of 24 in. Tantalum radiation shields surround the heater to reduce heat losses. Thermocouples are placed at known radial locations in the thick-walled tube so that both the heat flux and the inside wall temperature may be obtained from these temperature measurements.

Heat will be removed from the system by utilizing a thermal radiating coil condenser. The coil will be constructed from 1/2-in.-OD Nb-1% Zr alloy. Adjustable thermal radiation shields will surround the coil condenser to control the heat removal from the system.

The delivery of the vacuum chamber for housing the above facility is still delayed by the repair of leaks in the outside chamber cooling lines. The liquid nitrogen supply for the 10^{-8} -torr diffusion pump has been designed, and bids are being requested.

The design and selection of instrumentation is under study. The final thermocouple and pressure transducer designs will be completed during the next reporting period.

b. Heater Experiments.

(i) Thermal Radiation Heater Experiment. This experiment is being conducted to examine thermal radiation heat transfer further as a means of supplying high uniform heat fluxes and to get acquainted with the hardware used in this type of system. The loop has operated 40 hr since the thermal radiation shields surrounding the thermal radiation heated section were replaced. NaK temperatures ranging up to 1100°F have been obtained. Testing is underway to extend the present temperature level and heat flux range.

(ii) Electron Bombardment Heating Experiment. Assembly of the electron bombardment heated experiment is progressing satisfactorily. Clamshell heaters and insulation have been placed around the pool boiler. The system has been leak checked and found sufficiently leak tight.

G. Chemical Separations

1. Fluidization and Volatility Separation Processes

a. Recovery of Uranium from Low-enrichment Ceramic Fuels.

(i) Laboratory Work. Laboratory work in support of the pilot-plant investigation of the fluid-bed fluoride volatility process has continued by studying the fluorination of U_3O_8 - PuO_2 -fission product mixtures. This work is directed toward determining the optimum reaction conditions for the conversion of uranium and plutonium in spent uranium oxide fuels to UF_6 and PuF_6 , and minimizing the retention of plutonium on the alumina used as the inert fluidized bed material.

In previously reported work (see Progress Report for August 1964, ANL-6936, pp. 65-67), the fluorination of U_3O_8 - PuO_2 -fission product mixtures was carried out by a two-step procedure. Currently, tests are being carried out in a 1½-in. dia. fluid bed reactor to evaluate

a modified fluorination procedure wherein the entire charge of U_3O_8 - PuO_2 -fission product mixture is contained in the fluid bed at the start of the fluorination. This modification is required for fluorination of the material resulting from the chemical decladding of stainless steel-clad UO_2 fuel by HF-oxygen mixture (see Progress Report for July 1964, ANL-6923, pp. 68-69).

A series of tests were performed using the modified fluorination procedure to determine the effect of fluorine flow rate on plutonium removal from the alumina bed. In each test the reaction charge contained PuO_2 and fission products mixed with the alumina bed material. The fluorination reaction was conducted with 100% fluorine at three temperature levels: 5 hr at 450°C, 5 hr at 500°C, followed by 10 hr at 550°C.

In tests in which the fluorine flow rate was not sufficient to fluidize the alumina bed (100 to 300 cc/min), the plutonium concentration in the bed after fluorination averaged 0.074 w/o. Plutonium removal from the bed was significantly increased in tests in which the fluorine flow rate was sufficient to fluidize the alumina bed. At a fluorine flow rate of 4 liters/min, the alumina bed contained 0.022 w/o plutonium after fluorination. In tests employing 6 to 8 liters/min fluorine, the residual plutonium concentration was less than 0.004 w/o. These results indicate that a fluorine flow rate of 4 liters/min or less will be inadequate for efficient removal of plutonium from the alumina bed.

(ii) Decladding of Stainless Steel-clad Fuel. Development studies have continued on the decladding of stainless steel-clad uranium dioxide fuel by the high-temperature reaction of the cladding with oxygen in the presence of hydrogen fluoride (see Progress Report for July 1964, ANL-6923, pp. 68-69). A test was performed in a 3-in.-dia fluid-bed reactor to demonstrate both the destructive oxidation of the stainless steel and the fluorination of the products to recover uranium as uranium hexafluoride. The fuel bundle consisted of six 3-ft-long, type 304 stainless steel tubes (of 0.43-in. ID and 0.020-in. wall thickness) filled with cylindrical UO_2 pellets. The ends of the tubes were closed by stainless steel plugs. The UO_2 pellet charge weighed 4.5 kg, and the weight of stainless steel in the fuel bundle was 1.6 kg. The fluid bed consisted of 7.7 kg of sintered alumina particles.

Decladding was carried out in 3 hr by use of a reacting gas containing 43 v/o HF in oxygen. The bed temperature was maintained at 550°C for the first hour and at 450°C for the remainder of the decladding step. (A short circuit in a set of heaters caused the bed temperature to decrease to the lower value.)

After the decladding step, the entire bed, consisting primarily of fine particles of ferric oxide and uranyl fluoride with no trace

of the original metal cladding, was reacted with fluorine by a two-step procedure: 5 v/o fluorine in nitrogen at 400°C for 6.5 hr, followed by recycle fluorination at 450°C for 11 hr with the fluorine concentration gradually increasing from 5 to 90 v/o.

A somewhat unusual fluorine feed system was employed during the test to facilitate fluorination of the large quantity of uranium fines (40 w/o UO_2F_2 in alumina) in the bed. During the initial fluorination period, fluorine was introduced into the reactor at approximately the middle of the fluid bed and nitrogen as the fluidizing gas was fed at the bottom of the reactor. During the latter period, fluorine was fed with the fluidizing gas. Approximately 82% of the uranium was recovered as UF_6 during the initial fluorination period with UF_6 production rates averaging 10 lb/(hr)(sq ft). The alumina bed containing 25 w/o FeF_3 was found to be free flowing at the end of the run.

Work on the decladding of stainless steel-clad uranium dioxide fuel by hydrogen fluoride-oxygen mixtures is also being carried out at Brookhaven National Laboratory.¹⁴ It has been planned that optimization of the HF-O_2 decladding process will be conducted primarily at Brookhaven, and that work at Argonne will be concerned with the processing of declad fuel.

b. Recovery of Uranium from Highly Enriched Uranium Alloy Fuels

(i) Pilot-plant Demonstration Runs. An additional three runs have been made with unirradiated aluminum-uranium alloy fuel elements in the pilot-plant facility installed to demonstrate the recovery of uranium from highly enriched uranium alloy fuels by means of the fluid-bed fluoride volatility process (see Progress Report for July 1964, ANL-6923, pp. 70-72). During the hydrochlorination step, the volatile aluminum trichloride was converted to a solid oxide waste in a fluid-bed pyrohydrolysis reactor. Uranium hexafluoride product was collected on two sodium fluoride beds in series.

In two tests, successive charging of fuel elements was evaluated as a means for reducing processing time and for increasing HCl reaction rate and utilization efficiency. In each test, three elements were processed. The processing method involved charging of the fuel elements in a manner such that, at the time of charging, about 60% of the previously charged material had reacted with hydrogen chloride. In the demonstration runs in which three elements were processed successively, an increase of 20 to 30% in HCl utilization and average reaction rate was observed compared to values obtained in processing single-element charges. The hydrochlorination time per fuel element was reduced by approximately 25%.

¹⁴Brookhaven National Laboratory, Bimonthly Progress Report Volatility Process Development, July-August 1964.

The loss of uranium through the packed bed filter to the pyrohydrolysis reactor during the hydrochlorination step was significantly reduced by increasing the filter bed depth from 5-7 in. to 12 in., and by decreasing the bed particle size. With the deep bed, uranium losses averaged 0.6% of the uranium charge, compared with 3% loss with the shallow bed.

By employing a two-temperature fluorination procedure, more than 99% of the uranium was removed from the reactor bed material and more than 98.5% was removed from the filter bed, based on analyses of the bed materials after fluorination. This corresponds to a total uranium removal from bed and filter of about 99%. The fluorination procedure involved introducing dilute fluorine (6 v/o in nitrogen), with the reactor bed fluidized, at a temperature of 250°C and then slowly raising the temperature to 500°C over a 2- to 6-hr period. The fluorination was completed by operating with 30 to 60 v/o fluorine under static bed conditions for an additional 2 hr at 500°C. Recovery of the UF_6 product on the sodium fluoride traps was considered very satisfactory. In one test, 99% of the uranium charged to the reactor was recovered as UF_6 on the sodium fluoride traps during fluorination.

2. General Chemistry and Chemical Engineering

a. Studies of Mobile Blanket Fuels. The first material that will be seriously investigated as a mobile blanket material (see Progress Report for August 1964, ANL-6936, p. 68) will be a paste of uranium nitride suspended in sodium. A possible advantage of uranium nitride for this application is the anticipated compatibility of the UN-Na system (even at high burnup) with stainless steel container materials. Another advantage is that its uranium density and thermal conductivity are greater than those of uranium dioxide, which also was considered as a blanket material. Also, the difficulty of sintering uranium nitride should prevent drastic changes in its particle size when the blanket material is circulated in a reactor for long periods of time.

High-purity uranium nitride is being prepared. Meanwhile, uranium monophosphide, uranium monosulfide, and uranium dioxide are being used as stand-ins in order to develop techniques for measuring the physical properties of ceramic powder-liquid metal systems.

Work has continued on the measurement of wetting properties of ceramic blanket materials by liquid sodium. For a sodium-paste blanket fuel, an important consideration in the choice of a blanket material is the contact angle between sodium and the blanket material. Theoretical studies¹⁵ indicate that a small contact angle is desirable. In scoping runs,

¹⁵J. Woodrow, H. Chilton, and R. I. Hawes, J. Nuclear Energy, Part B, 1, 229 (1961).

the contact angle for sodium or uranium monophosphide has been measured at about 260°C and was found to be 130°; the contact angle for sodium on uranium monosulfide at about 245°C has been found to be about 150°.

Preliminary studies, in which pastes were made of UO_2 powder and either low-melting bismuth alloys or sodium, have indicated the importance of the physical properties of the solid particles in determining the characteristics of the paste. Specifically, UO_2 of 1.96-g/cc bulk density could be dispersed in low-melting bismuth alloys or in sodium to the extent of only 18 to 20 v/o, whereas a 50 v/o UO_2 sodium-suspended paste could be made from UO_2 of 5.37-g/cc bulk density.

b. Preparation of Uranium Monocarbide. Work has continued on a method to produce uranium monocarbide (UC) by hydriding and dehydriding uranium metal to form powder, which then is reacted with a $\text{C}_3\text{H}_8\text{-H}_2$ mixture in a fluidized bed. The most recent run, performed in a stainless steel column, utilized a superficial gas velocity of 0.75 ft/sec, a gas composed of 79 v/o $\text{C}_3\text{H}_8\text{-21 v/o H}_2$, a temperature of 580°C, and a run time of $5\frac{1}{3}$ hr. The total carbon content of the product was 4.45 w/o (stoichiometric UC contains 4.80 w/o carbon). The presence of UC in the product was verified by X-ray analysis. Additional runs will be made at a reduced fluidizing gas velocity (and thus a decreased entrainment of fine uranium particles in the cold zone of the column) in an attempt to lower the reaction time and to increase the carbon content of the product.

H. Plutonium Recycle Program

1. EBWR Plutonium Recycle Experiment

One of two fuel storage racks has been fabricated and delivered to the EBWR. The second rack will be completed early next month. These racks will be used for storing the core associated with the Plutonium Recycle Program. The original fuel-storage racks have been removed from the pit to make room for the 1.4% boron-stainless steel racks.

2. Plutonium Recycle Fuel

An additional quantity of 400 central-type Plutonium Recycle fuel rods (beyond the 810 already received; see Progress Report for August 1964, ANL-6936, p. 72) are through final inspection at Hanford and ready for shipment to ANL. Completion of the remaining 300 fuel rods has been temporarily delayed pending Pu decontamination of welding facilities.

In addition to the regular $\text{PuO}_2\text{-UO}_2$ fuel for the central zone, Hanford has started preparation and loading of 25 additional test fuel rods which will be used for physics measurements on the core.

Can stock needed by United Nuclear Corporation for the enriched and natural fuel element loadings has been formed, and preparation of the evaluation fuel cans has been completed. Approximately 75% of the Zircaloy-2 jacket tubing has been processed through the final inspection and accepted for loading. Approximately 366 m (1200 ft) of tubing fabricated by the subcontractor for United Nuclear was diverted by the subcontractor for another order. Fabrication of additional tubing to replace this material has been started.

A nondestructive testing overcheck by ANL on approximately 3% of each lot of tubing delivered to United Nuclear has revealed one rejectable crack-type defect in one tube from Lot No. 29.

Evaluation lots of natural UO_2 pellets, enriched UO_2 pellets containing burnable poison, Zircaloy-2 end plug stock, Zircaloy-2 jacket tubing, finish machined end plugs, stainless steel retainer springs, stainless steel upper support grids, stainless steel lower end fittings and 144 evaluation fuel rods loaded with natural UO_2 pellets have been delivered to, inspected, and accepted by ANL. Upper support grids for plutonium fuel rods and stainless steel lower support grids have been rejected as not conforming to specifications and will be replaced. Fuel-element cans have not been received for evaluation.

All the natural UO_2 pellets and all the 6% enriched UO_2 pellets containing the rare earth burnable poison have been fabricated, tested and inspected. These pellets have been assembled into weighed stack-heights ready for loading into fuel jacket tubing.

3. Plutonium Recycle Control Rods

A total of eight new control rods and one spare central oscillator rod are being fabricated for the Pu Recycle Experiment. The eight control rods have boron-stainless steel absorber sections with 2 w/o boron enriched to 90 w/o B^{10} contained in Type 304 stainless steel. The absorber section of the oscillator rod consists of hafnium.

The boron-stainless steel was fabricated by Carpenter Steel Co. in accordance with ANL specifications. A total of 34 pieces of strip stock were finished to 0.33 x 28 x 150 cm (0.130" x 11" x 60"). The total boron content analyzed 1.92 to 1.94 w/o for strip from Heat No. V-91077 and 1.86 to 1.87 w/o for strip from Heat No. V-91084.

All hafnium for the oscillator rod was converted to plate stock by Wah Chang Corporation from material furnished by ANL.

Completed control rods and oscillator rod are being fabricated by Dresser Products, Inc., Great Barrington, Mass., in accordance with ANL specifications. All boron-stainless steel angles required for the absorber

cruciform sections have been formed, spot welding procedures evaluated and approved, and one cruciform section completed. All Zircaloy-2 and stainless steel stock required for the follower sections and cross guide followers has been formed and is ready for spot welding. Most of the remaining miscellaneous components required for control rod assembly and the hafnium plate stock for the oscillator rod have been finish machined.

The complete assembly of all control rods has been scheduled for October 23, 1965.

4. Nondestructive Tests of Components for the Plutonium Recycle Experiment

a. Inspection of Zircaloy-2 Tubing. Originally, ultrasonic techniques were to be applied to the evaluation of loaded plutonium recycle tubing only. However, it has since been decided to check four unloaded tubes, randomly selected, from each lot. Unloaded tubes from early lots were received at Argonne during the first week of August.

Testing is being accomplished on a stuffing box system that consists of a drive roller assembly, stuffing box, and an idler roller assembly. Each tube is passed through the system four times: twice for longitudinal defects and twice for transverse defects. In the longitudinal and transverse tests, the tubes are run in both directions to insure detection of unfavorably oriented defects. A 5-Mc, spherically focused lithium sulfate transducer is employed in both tests. The inspection system is calibrated by means of standard tubes which contain series of ID and OD longitudinal and transverse electromachined notches. As of this writing test results are as follows:

(i) Lots 4 and 18 contained tubes which appeared to have transverse defects from 0.051 to 0.076 mm deep. Subsequent metallography revealed that these defect indications were actually caused by small deposits of metal occurring at the end of long, shallow scratches caused by the air gauging of the tube ID. In all instances, borescopic examination revealed the presence of this buildup in suspicious areas. As an added precaution, these tubes were subjected to an eddy-current inspection, which confirmed that these defects were in fact small areas of buildup metal.

(ii) One tube in Lot 28 has an ID longitudinal defect that appears to be deeper than 0.051 mm. This defect was also detected with eddy currents and a borescope.

5. Experimental Program

A detailed experimental program has been developed. First, the critical mass of the unpoisoned plutonium-zone fuel will be measured; the remaining elements of the 36-element plutonium zone will then be

loaded into the reactor vessel and boric acid will be added to the moderator to maintain criticality. Boric acid worth, temperature coefficient, transfer function, and foil-activation experiments will then be made to check the accuracy of the calculational model.

Next, the shim and natural uranium zones will be loaded. Control rod worths, boric acid worth, and temperature coefficients for the full system will be obtained; transfer measurements will be performed during the approach to power to determine the highest power level at which stability can be maintained. The reactor will be operated at this power level until the plutonium-zone fuel has received an exposure of 2000-3000 MWd/Tonne. The reactor will then be shut down, the shim and natural zones will be unloaded, and the sequence of experiments will be repeated.

6. Physics Calculations

The calculational model previously described (see Progress Report for July 1964, ANL-6923, pp. 75-8) has been used to check the experimental data obtained from Hanford's subcritical experiments. The fuel consisted of rods having a composition of 1.2 w/o Pu-Al and which were 0.635 cm in radius, 111.3 cm in length, and clad with 0.0762-cm-thick zirconium. The fuel rods were arranged in a triangular pattern. Five lattices were examined with H/Pu atom ratios ranging from about 630 to 1420.

The isotopic composition of the plutonium was of 93.97% Pu^{239} , 5.58% Pu^{240} , and 0.44% Pu^{291} . The aluminum in the rod was considered to be a moderator for the purpose of evaluating the resonance absorption in Pu^{239} and Pu^{240} . Table XII lists various calculated results.

Table XII. Calculated Lattice Parameters for 1.82 w/o Pu-Al Lattices

H/Pu Atom Ratio	Pu^{240} Resonance Integral, b	Fuel Disadvantage Factor	$\bar{\eta}f$	Error, % δk_{eff}
632.5	6490	1.091	1.506	1.2
812.8	6580	1.095	1.445	1.2
1004.5	6630	1.099	1.384	0.9
1208.1	6670	1.104	1.323	0.8
1423.3	6690	1.108	1.264	0.6

The error in the calculated k_{eff} value is larger than ones previously considered in oxide systems. The increase in error may be due to the lack of U^{238} in the Pu-Al system coupled with the possible overestimation of the U^{238} resonance integral.

A new 3600 code, SAMPLE (RP 352X), is being written to study the isotopic variation in isolated materials imbedded in a spectrum determined

by the CYCLE 2 code (RP 282). This code will be of use, for example, in the study of the special fuel pins to be irradiated and periodically analyzed in the EBWR plutonium zone.

In anticipation of this code, CYCLE 2 has been modified so that it will now, at user option, prepare input to be used by SAMPLE. This input takes the form of group fluxes and integrated flux-time data from each burnup step of the CYCLE 2 problem. The data is written in binary coded decimal on tape 8.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Fuel and Core Design

Phase I of the fuel-fabrication-process verification studies conducted by the Martin Company, Battelle Memorial Institute, and Pyromet Company has been completed.

The work by BMI on fabrication of cermet fuels (37 w/o UO_2 -SS) by compaction and rolling of loose powder compacts has produced optimum homogeneity of dispersion with minimum fragmentation of a nominal-strength oxide by means of careful control of essentially three factors: (a) blending process, (b) compaction schedule, and (c) rolling schedule at proper temperature.

Evaluation of various blending processes has shown that the best homogeneity is achieved with wax-coat and fine stainless steel-powder buildup over the oxide particles. The camphor-alcohol and powder mixing-blending technique, and the paraffin- CCl_4 and powder mixing-blending technique yielded poorer homogeneity. Minimum fragmentation of the nominal-strength oxide was achieved by using 6:1 reduction of the compact in a rolling schedule which used heavy reductions in the initial passes and lesser reductions in the final stages of rolling. Optimum quality was achieved on the 8-in.-diameter rolling mill by using a schedule of 40, 40, 25, 25, 25, etc., reduction per pass on compacts heated to 1200°C .

The work by Martin Company to adapt the advanced army reactor fuel fabrication process to the AARR fuel design has met with success. Verification-proof plates have demonstrated that forming of the SS coated-oxide particle cermet in the green state by means of die extrusion can indeed yield full size flat plates.

Pyromet Company has demonstrated that the rhomboidal AARR fuel assembly can be assembled and brazed to meet close dimensional tolerances with reasonable care and design of the brazing fixtures. The problem of 100% quality assurance for all brazed surfaces has not met with complete success. The solution appears to lie in the method of application of the brazing compound and the selection of optimum furnace operating conditions.

2. Reactor Component Design Studies

The research and development contract first reported in the Progress Report for May 1964 (ANL-6904) has been satisfactorily concluded.

This contract provided some of the design parameters necessary for the reactor and for the reactor system. Data and design have been furnished on the following tasks:

- a. conceptual design and verification of stress calculations;
- b. reflector stress analysis;
- c. nuclear instrumentation study;
- d. process control stability study;
- e. fuel-transfer system design study;
- f. control rod drive mechanism study.

Neutron and gamma flux distributions were computed in order to determine the stresses and cooling requirements in the beam tubes and in the reflector. Information relating to the design of the reactor vessel was produced during the course of these tasks and in the course of the nuclear instrumentation study. The nuclear instrumentation study supplied the placement as well as the details of operation at various radiation levels of the reactor nuclear instrumentation. Reactor kinetics were used to augment an analog study involving the response of the process control system to changes in reactor operating power level. The feasibility of close control despite long process delay times was clearly indicated and the principal component design values involved were evaluated. A conceptual design of the fuel transfer and of the control rod drive mechanisms were made.

B. Magnetohydrodynamics (MHD)

1. MHD Power Generation - Jet Pump Studies

The data gathered during previous studies of the jet pump MHD system operating on steam and water have led to several conclusions. Very good efficiency can be realized in the conversion of enthalpy to kinetic energy in the steam jet. However, considerable degradation of available energy is encountered in the combiner and condensing region. Also, for the velocities involved, the hydraulic losses are reasonable. These observations indicate the necessity for concentrating development upon the combiner region of the jet pump. Presently, this consists of exploring the efficiency capability of this unit by construction and operation of a unit with variable aspect ratio. This will permit optimization of the operating geometry with possible significant increase in efficiency.

2. MHD Studies with Liquid Metals

Extensive analysis of the three principal cycles that have been proposed for liquid metal MHD power systems has been completed. The three cycles studied were the two-component, two-phase cycle, the condensing injector cycle and the one-component, two-phase cycle. The three cycles are basically similar in that they are based on the conversion of thermal to kinetic energy or stagnation head, which is then converted into electrical energy by an MHD device. The cycles differ primarily in the manner in which the conversion of thermal energy is achieved. The objectives of the cycle studies were to investigate and establish the potential of the liquid metal MHD concept and to make a preliminary judgment of the relative merits of the cycle as applied to commercial power systems. The working fluids studied were mercury-potassium alloy, potassium, cesium, and sodium. The heat source and sink temperatures were varied to correspond to conditions that are compatible to pure and topping MHD cycles.

The following are typical pertinent conclusions that evolved from these studies:

(1) The two-phase, single-component and condensing injector cycles have essentially the same cycle efficiency for a specified temperature range. The efficiency of the two-component, two-phase cycle is lower by a factor of approximately two and therefore appears to be the least attractive for commercial application of the cycles studied. The lower efficiency of the two-component, two-phase cycle is traceable to the large kinetic energy losses in the separator and the additional power requirements needed for the vapor loop EM pump.

(2) The maximum overall efficiency obtainable for a liquid metal MHD topping and steam bottoming cycle ranges between 55 to 60%, and results from using either potassium or cesium as the working fluid. The liquid metal topping cycle operates between a temperature range of 2240 and 1100°F, and produces 40% of the total power.

(3) The maximum overall cycle efficiency is generally reached when the bottoming steam cycle is operated at maximum allowable temperature conditions. As the sink temperature is raised, the decrease in the efficiency of the topping cycle is more than offset by the increase in the steam cycle efficiency which results in a maximization of the overall cycle.

(4) The maximum efficiency for a mercury-steam binary cycle is approximately 54%, which is slightly less than that of the cesium or potassium steam binary cycle. This is based on a source temperature of 1540°F

and condenser temperature of 1100°F. If the sink temperature is dropped to 540°F, the overall cycle efficiency drops to 47% even though the efficiency of the topping cycle increases.

(5) The working fluid which appears to be most attractive for a pure MHD power system is mercury, because mercury is superior from the thermodynamic viewpoint and can operate in the lowest temperature range. The maximum efficiency of such a pure MHD cycle would be 28% operating between the temperature limits of 1540 and 450°F. Such a cycle may be quite attractive from an economic viewpoint in spite of the relatively low efficiency.

The attractiveness of the pure MHD power cycle results from the major simplification that could be achieved in the overall plant with its resultant economies.

(6) Regardless of the temperature range specified, the potential overall cycle efficiency of a liquid metal MHD-steam binary cycle is superior to the plasma MHD-steam binary cycle. When the temperature ranges are taken into consideration, the attractiveness and superiority of the liquid metal MHD cycle is enhanced even further.

Because of several major uncertainties that can only be resolved through experimentation, it is impossible at present to clearly establish the superiority of either the condensing injector cycle or the two-phase, single-component cycle.

C. Regenerative EMF Cells

1. Bimetallic Cells

a. Sodium-Bismuth Cells. The ternary eutectic NaI (54.6 m/o)-NaCl (28.7 m/o)-NaF (16.7 m/o) is being studied for possible use as the electrolyte for the sodium-bismuth cell. Since a high solubility of sodium in the electrolyte would possibly lead to an irreversible sodium transfer from anode to cathode, the solubility of sodium is under investigation. Preliminary determinations (see Progress Report for August 1964, ANL-6936, p. 76) of the solubility of sodium in this electrolyte were thought to be low because of sodium losses by volatilization.

Additional solubility experiments were performed in which sodium volatilization was reduced by limiting the gas volume above the melt. Agreement with previous results indicates that loss of sodium by volatilization was not a significant factor in the initial experiments. A single determination of the solubility of sodium in the binary eutectic NaI (64 m/o)-NaCl (36 m/o) agreed with solubility data for the ternary eutectic.

2. Lithium Hydride Cell

A knowledge of the thermodynamic activity of LiH in fused salt systems is important in evaluating a lithium-hydrogen direct energy conversion device. The determination of the standard emf of the lithium hydride cell,

(see Progress Report for May 1964, ANL-6904, p. 101) made possible the calculation of the activity of LiH in similar cells. By use of these data, for example, it is expected that the regeneration temperature of the system can be established.

The effect of electrolyte composition on the activity of LiH is being investigated. Because previous determinations for LiH in the LiCl-LiF eutectic gave higher values of the activity than expected (see Progress Report for August 1964, ANL-6936, p. 76), studies are being directed toward the determination of the activity of LiH with only LiCl as the electrolyte. As a basis for comparison, activities of LiH in both electrolytes have been calculated from emf measurements at 610°C. For 11.01 m/o LiH and 5.64 m/o LiH in the LiCl-LiF eutectic, activities were calculated to be 0.34 and 0.19, respectively; for 10.20 m/o LiH and 1.04 m/o LiH in LiCl, activities were calculated to be 0.26 and 0.026, respectively. Examination of these data indicates that LiH exhibits strong positive deviation from ideality in LiCl as well as in the LiCl-LiF eutectic.

V. NUCLEAR SAFETY

A. Thermal Reactor Safety Studies

1. Metal-Water Reactions

a. Studies in TREAT of Uranium Dioxide Fuel. Studies were continued of the reaction of water with oxide-core, Type 304 stainless steel-clad fuel pins when the pins, immersed in water, are exposed to TREAT transients. The objective of these studies is to obtain kinetic data under conditions that simulate a power excursion or a runaway incident. Studies with Zircaloy-2-clad fuel pins were summarized in the Progress Report for June 1964, ANL-6912, p. 90. The equipment and experimental conditions described therein were also used in the experiments with stainless steel-clad pins discussed here.

The extent of metal-water reaction was computed, as in previous studies, from the amount of hydrogen evolved, considering stainless steel to be equivalent to iron and using the following stoichiometry:

Particle size determinations of the fragmented fuel were made by a sieve-screen analysis.

Experiments have been performed with pure UO_2 -core fuel pins and with mixed oxide-core fuel pins. Two experiments were with UO_2 -core pins which had been waterlogged. (Waterlogging was accomplished by drilling two small holes, one in each end of the fuel pins, and soaking the pins in water. The drilled holes were subsequently filled with stainless steel wire.) Experimental results are summarized in Table XIII and Figure 27. In Figure 27 each datum is located by its extent of reaction and by its fission energy input (cal/g of core). The number beside each point is the mean volume-surface particle diameter of the core and cladding material as determined by sieve-screen analyses. In order to place the experiments with the mixed oxide-core pins on the same basis as the UO_2 -core pins in Figure 27, two energy scales and one temperature scale are used in the figure. The energy scales were placed so as to give correspondence with the single temperature scale by choosing a melting point of 2700°C as the fixed point of comparison.

It is of interest to compare the results of experiments with stainless steel-clad fuel pins with those reported for the Zircaloy-2-clad pins in ANL-6912.

Table XIII. Treat Metal-Water Meltdown Experiments with Oxide-core, Type 304, Stainless Steel-Clad Fuel Pins

(Water initially at room temperature) (Initial helium overpressure of 20 psia)							
Fission Energy Input (cal/g)	Adiabatic Core Temp (°C)	CEN Run No.	TREAT Characteristics		Appearance of Specimen after Transient	Mean Particle Diameter (mils)	Percent of Metal Reacted
			Period (msec)	Integrated Power (MW - sec)			
Mixed Oxide Core Fuel Pins							
(Cladding: SS-304, 20 mils thick, ends capped. Core: 81.5 w/o ZrO ₂ , 9.1 w/o CaO, 8.7 w/o U ₃ O ₈ , fully enriched, 0.74 w/o Al ₂ O ₃ , 0.33 in. dia, 0.50 in. long)							
296	1900	40	60	315	Intact	330	0
395	2500	21	78	420	Intact	330	0
432	2600	48	72	460	Clad Ruptured/ Core Fragmented	15	4.3
536	3200	24	65	560	Clad Ruptured/ Core Fragmented	-	6.6
UO ₂ Core Fuel Pins							
(Cladding: SS-304, 20 mils thick, ends capped. Core: UO ₂ , 20% enriched except runs 169 and 174, 11.2% enriched, 0.33 in. dia, 0.50 in. long)							
152	2000	63	121	120	Intact	330	0.3
215	2700	67	290	185	Intact	330	0.3
242	2700	65	50	190	Fragmented	4	5.2
292	2900	66	97	230	Fragmented	23	9.0
312	3100	69	64	245	Fragmented	2	5.4
407	3300	61	116	320	Fragmented	17	19.1
432	3300	62	49	340	Fragmented	13	14.0
437	3300	60	47.5	344	Fragmented	11	13.5
510	3300	71	42.5	400	Fragmented	8	11.5
617	3300	169	42	768	Fragmented	26	29.8
655	3300	170	51	606	Fragmented	10	31.7
688	3300	174	39	860	Fragmented	10	35.5
(Core Saturated with Water before Test)							
97	1300	72	110	76	Clad Ruptured	330	0.7
178	2300	64	115	140	Clad Ruptured	330	0.8

Figure 27. Results of Meltdown Experiments in TREAT with UO₂-core and Mixed Oxide-core Type 304 Stainless Steel-clad Fuel Pins Submerged in Water

(1) In each set of data, there was an approximate line or relationship between the extent of metal-water reaction and the fission energy input. For the stainless steel cladding, the threshold for significant metal-water reaction coincided with the beginning of UO_2 melting, but the threshold was lower for the Zircaloy-2 cladding (about 1600°C adiabatic core temperature). In the energy range corresponding to nearly complete vaporization of UO_2 , the stainless steel-clad pins reacted to a maximum of 35% while the Zircaloy-2-clad pins reacted to as much as 80%. The extent of reaction in both cases was nearly independent of the nature of the core material (i.e., UO_2 or mixed oxide).

(2) The energy threshold for fragmentation does not appear to be completely independent of the nature of the core material. Both stainless steel-clad and Zircaloy-2-clad UO_2 -core fuel pins showed an energy threshold for fragmentation of about 240 cal/g of fission energy input (adiabatic core temperature 2700°C , partly melted). The energy threshold for fragmentation of the mixed oxide-core fuel pins with both types of cladding was somewhat lower than that of the UO_2 -core pins. Cladding rupture and partial dispersion of the mixed oxide-core material occurred at an adiabatic core temperature of 2600°C for stainless steel-clad pins (CEN-48); similar ruptures occurred at adiabatic temperatures as low as 1900°C for Zircaloy-2-clad pins. It should also be noted that in experiments with waterlogged stainless steel-clad UO_2 -core pins, cladding ruptures occurred at very low fission energy inputs (CEN-64 and 72).

(3) In each set of data, pressure transducer recordings showed that both the peak pressure in the autoclave and the rate of pressure rise increased as the energy of the reactor pulse was increased. No explosive pressure increases were observed in tests of fuel pins with either type cladding. For stainless steel-clad pins, the most rapid rate of pressure increase was about 6 psi/msec (CEN-170); for Zircaloy-2-clad pins, it was about 1.5 psi/msec (see results for CEN-173 in Progress Report for June 1964, ANL-6912, p. 92).

2. Metal Oxidation-Ignition Studies

a. Ignition Studies of Irradiated Uranium. The program to determine the effect of irradiation on the ignition behavior of uranium has been completed. As a part of this program, the effects of thermal cycling (to simulate thermal exposure from fission heat) and of background radiation on the ignition behavior of uranium were also investigated. Samples of 20% enriched uranium in the form of right cylinders (6.4 mm x 5.1 mm in dia; specific area, $0.60 \text{ cm}^2/\text{g}$) were used for all the experiments. A small sample size (high specific area) was chosen so that the ignition temperature would be expected to be no greater than 600°C , where post-irradiation swelling is known to occur. During irradiation, samples were enclosed in

constrainment jackets and maintained at modest temperatures ($< 230^{\circ}\text{C}$) to avoid dimensional changes and swelling. Ignition experiments both in air and in oxygen were performed with control samples, thermocycled control samples, and samples irradiated in MTR to burnups of 0.8 and 1.7% of the uranium-235.

It has previously been reported (see Progress Report for July 1964, ANL-6923, p. 87) that thermal cycling did not produce any change in ignition temperatures of uranium in air or in oxygen. Burning curve ignition experiments conducted in air and in oxygen in the presence of a high radiation background showed no change in ignition temperatures as a result of the background radiation.

Samples which were irradiated to 0.8 and 1.7% burnup of the uranium-235 showed ignition temperatures in oxygen of 400°C and 360°C , respectively, as compared with 600°C for nonirradiated material. When the irradiated fuel was heated in air, no definite ignition was observed. However, at about 350°C , greater self-heating occurred in the irradiated samples than in the corresponding control samples. From these experiments it can be concluded that (1) in oxygen, a marked decrease in the ignition temperature of uranium results from irradiation, and (2) in air, an increase in the oxidation rate of uranium results from irradiation and this increase very likely leads to a decrease in ignition temperature.

B. Fast Reactor Safety Studies

Two of the most important factors influencing the safety characteristics of fast power reactors are the large quantity of fuel and the short prompt-neutron lifetime. Because of these, it is possible to postulate a class of "meltdown and reassembly" accidents consisting of three stages as follows: (1) some operating abnormality occurs which produces some degree of fuel failure; (2) material movements occur, resulting in a more reactive configuration; and (3) a destructive burst of nuclear energy terminates the accident. The mechanisms involved in fuel failure, reassembly of fuel into supercritical mass, coolant effects, etc., are complex and not well understood. Consequently, these mechanisms are being studied experimentally and analytically in a program centering on tests being performed in the TREAT reactor.

1. Preliminary Studies for Loop Experiments in FARET

The first meltdown experiment was performed in a small sodium loop in TREAT. Development effort is well along on a large sodium loop designed to be a semipermanent installation in TREAT (see Sect. VC). Preliminary studies are now being made on loop experiments to follow.

Although TREAT is a thermal reactor and has a minimum reactor period of 40 msec, it is still basically appropriate for study of meltdown-type accidents, since such incidents are typically considered to have time scales of the order of seconds or fractions of seconds. However, the thermal-neutron flux depressions in TREAT can be a real limitation in studies on large clusters of sample fuel elements. It is possible to minimize power variations from sample to sample in a cluster by varying the enrichment in the samples. This procedure, however, does not correct for radial or circumferential power distortions inside individual samples, and may actually enhance the spatial variations in power once the fuel has failed and begins moving. Thus, TREAT is fundamentally a tool for studying phenomena occurring in small clusters.

Although FARET will not be a transient test facility, it will have a fast-neutron spectrum which will make it possible to perform closed-loop experiments with large clusters of fuel pins under representative spatial power distributions. Consequently, it is possible to study the behavior of large clusters of fuel elements run to failure under or near steady-state conditions, and simulated pump-failure accidents. A closed-loop facility could also be used to study, safely and conveniently, the behavior of vented fuels and the release of plutonium or fission products from failed elements.

The current design status of FARET has been reviewed to determine the feasibility of including a closed loop, similar to the large TREAT sodium loop, but having the capability of running steady-state destructive tests on samples. Conclusions of the study are:

- a. A closed loop is feasible and compatible with the dimensions of the current design.
- b. The pipe opening in the center of the reactor vessel cover for the loop should be larger than the 3-in. Schedule 80 piping specified for control rods. However, there is sufficient room for an opening of adequate diameter.
- c. The loop can be readily installed in and removed from the reactor vessel if one of the eight control rod drives is removed.
- d. There appears to be adequate room for proper placement of components and for bringing the loop into the reactor cell through the access tunnel.

2. Modifications of TREAT Integral Sodium Loop

All operational phases of the first TREAT experiments with the first integral sodium loop have been completed and have provided an empirical basis for review of possible modifications of the facility for future meltdown

experiments. The first loop was loaded with a single EBR-II prototype fuel pin previously described (see Progress Report for July 1964, ANL-6923, p. 88). The utility of the current design and procedures has been fully demonstrated. However, significantly increased capability for re-use and higher performance are expected to result from the following changes:

a. Loop design will be modified to permit filling, drawing, and flushing of the loop. This change should facilitate removal of sample fuel fragments following experiments in which extensive sample failure and dispersion occurs, as well as some handling operations.

b. Special high-temperature transformers for the ac electromagnetic pump will be fabricated and used to replace the present transformers. The change will permit the equilibrium temperatures of the sodium in the loop, prior to the transient test, to be increased from its present limit of less than 300°C. A sodium operating temperature of 500°C is desired.

Additional changes have been made in electrical connections, heater configurations and installation, and flowmeter installation to reduce the time required to prepare the three remaining loops. These changes should also decrease, if not eliminate, damage to leads during loop disassembly.

Limitations on the maximum stable flow rate, which is now less than one-half the desired 28 ft/sec velocity are being investigated in an attempt to obtain flow comparable with design EBR-II conditions.

C. TREAT

1. Large TREAT Loop

The integrity of the shielding in the newly built cask was determined by the technique of measuring the attenuation of γ radiation supplied by a known high-level source. The shielding appears to be adequate and the attenuation is close to the calculated value. A small amount of shop work had to be performed upon the lower seal ring to permit positive sealing. The cask will be used to ship an irradiated fuel assembly from the TREAT site to Argonne, Illinois.

Another shipment of parts (which will be used in loop construction) was sent to Idaho. This equipment included the 2- and 1-in. Fulton Sylphon bellows sealed valves, plugging indicator with heat exchanger, pipe-support parts, and flow meters.

Hydraulic calculations indicate that the flow losses in the primary system downstream from the header will be less than the head of sodium that can be maintained in the downcomer pipe. It is not expected that this pipe will be completely filled with sodium during loop operation, obviating the need for a siphon-breaking device.

The design of the gas system has been completed with the addition of two gas lines, to vent the inlet and outlet header, and the surge suppressor. A rearrangement of the piping in the Sodium Equipment Room and addition of regulation and control devices were made necessary by the relocation of the helium-gas supply to the reactor floor. The loop-insulation specification has been completed. Local contractors have been invited to make bids.

The electric heaters for the tanks were received from the manufacturer, attached to the tanks, and bonded with heat transfer cement.

Welding of piping subassemblies that do not require expansion joints was completed. Delivery of the expansion joints is being delayed because the manufacturer is having difficulty making acceptable welds. It now appears that the starting date for installation of the loop equipment in the Sodium Equipment Room will be determined by the delivery of the expansion joints. Prefabrication of the pipe and valve supports is in progress, and about 75% of these supports have been completed.

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