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Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

January 1966

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Reactor Technology
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AEC Research and
Development Report

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

January 1966

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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 overall 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:

July 1965	ANL-7082
August 1965	ANL-7090
September 1965	ANL-7105
October 1965	ANL-7115
November 1965	ANL-7122
December 1965	ANL-7132

REACTOR DEVELOPMENT PROGRAM

Highlights of Project Activities for January 1966

EBR-II

EBR-II has now been run to a total integrated power of 5570 MWdt. Run No. 14 was completed January 23 and the reactor was then shut down for a planned 9-week shutdown period to allow modifications to be made and to conduct the required annual tests.

The minor chemical explosion in December 1965, due to ignition of hydrogen and oxygen formed during charging of the interbuilding-coffin batteries, damaged essentially only the inner door of the equipment air lock. Revised procedures for handling of the interbuilding coffin to avoid such an incident are now in effect.

ZPR-9

Studies with Assembly No. 9 have been completed. Assembly No. 8 was constructed to study the properties of a beryllium oxide-reflected tungsten based core. Criticality was achieved with a fuel inventory of 283.9 kg U^{235} .

Plutonium Recycle Program (EBWR)

The moderator temperatures coefficient for the 36-assembly loading with boric acid concentrations in the neighborhood of 5 g/gal were remeasured.

AARR

Critical experiments with the core containing 810 fuel-foils and 810 boron-stainless steel poison strips have been completed. A new core containing 1215 fuel foils (~ 62.1 kg U^{235}) and 1620 boron-stainless strips (850 g boron) was brought to criticality.

Reactivity-control worths of the control blades were measured. It does not seem that stainless steel followers for the control blades can be used because of too great a reactivity loss.

A Selection Board has been established to select a manufacturer for the AARR reactor vessel.

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I. LIQUID-METAL-COOLED REACTORS

A. EBR-II

1. Operations

Integrity of the equipment air-lock vessel and outer door, which has been involved in the chemical explosion in December (see Progress Report for December 1965, ANL-7132, p. 1), was confirmed by hydrostatic and air-pressure testing. The test head which is normally used for leak-rate testing of the air lock was used to seal the inner opening in place of the door which had been damaged. This test head is also being used for normal traffic through the air lock until the replacement door is installed.

With verification that the reactor containment building, the reactor, and its auxiliary equipment were unimpaired, Run No. 14 was begun and finished, adding 520 MWdt of reactor operation, bringing the total integrated power to date to 5570 MWdt.

The reactor was then shut down as planned for modifications to systems and the required annual tests of the plant. This shutdown will last approximately nine weeks. Work to be performed includes replacement of 200 dip ring heaters in the large and small rotating plug seals, replacement of six primary tank immersion heaters and their control system, and replacement of Control Rod Drive No. 9. Numerous miscellaneous jobs can best be done when the plant is shut down will also be performed during this period.

2. Chemical Explosion of December 1965

A detailed investigation has been made of the conditions leading to the chemical explosion which damaged the equipment air-lock door (see Progress Report for December 1965, ANL-7132, p. 1). The explosion was traced to the accumulation and ignition of hydrogen and oxygen liberated from the interbuilding-coffin batteries which had inadvertently been left on a high charging rate.

The pressure developed by the deflagration of the gas in the air lock was calculated to be not greater than about 50 psia before relief was obtained by the lifting of the inner door. These calculations were based on overcharging of the batteries with a concurrent production of about 400 cu ft of hydrogen and 200 cu ft of oxygen.

The forces developed were estimated to produce a stress of about 1200 psi at the welded junction of the air-lock cover extension. This low value of stress was insufficient to cause any damage to the structure. Essentially, the only damage to the air lock was to the inner door, which will be replaced.

In spite of the low value of the calculated stress, all available air-lock welds were visually inspected with a magnifying glass to verify that the equipment air lock was undamaged. After all inspections were completed, the equipment lock was filled with water and tested at 30 psig hydraulic pressure at the highest point in the air lock. No leaks or defective areas were found. The lock was then emptied, and a pneumatic test of 22-hr duration at 36.2 psia was performed. The leak rate was found to be 52 cu ft for a 24-hr period, which is well below the leak rate of 70 cu ft measured previously and which is considered acceptable.

Revised procedures for handling of the interbuilding coffin have been insured and are in force. The major provision of the revised procedures is to limit the residence time of the interbuilding coffin in the air lock only to the short periods required for transfer of the coffin between the Reactor Building and the Fuel Cycle Facility. Also, the electrical circuit was revised to preclude battery charging in the air lock.

3. Reactor Improvements

a. Improved Pump for Secondary System. The purchase contract for the improved AC Linear Induction Electromagnetic Pump for the Secondary System (see Progress Report for September 1965, ANL-7105, p. 2) was awarded to General Electric Company on November 10, 1965. Since that time the manufacturer has been engaged in the design and engineering of the pump, primarily in optimization studies to determine the best hydraulic and magnetic arrangement which will eliminate the duct vibrations that caused fatigue cracks in the original pump. Procurement of materials for the pump is scheduled in February with delivery of the unit in Idaho by September 1966.

b. Additional Superheater Units. Requests for quotations for additional superheater units for the EBR-II Steam Generator were submitted to manufacturers in December 1965. Proposals are to be returned by February 11, 1966. The specification for the superheater units requires that the manufacturer supply one unit with an option for an additional unit, both of which would duplicate the existing superheaters at EBR-II. These new units would employ the same "modified evaporator" design as used previously. Although the design is fixed, the manufacturer will be allowed to employ procedures in construction and testing which differ from those used during the construction of the original units, subject to review by ANL.

c. EBR-II Digital Controller. A more detailed description of each of the six major components of the digital controller (see Progress Report for October 1965, ANL-7115, pp. 6-7) and some of their operating characteristics are as follows:

(i) The analog-to-digital signal conversion system consists of the following:

(a) A long-life, low-speed, reed relay multiplexer which places the values of twenty guarded-differential input signals into twenty amplifiers (one signal per amplifier). This multiplexer contains eight individually addressable channels, each with 20 parallel analog signals, making a total of 160 possible analog signal inputs to the digital controller. However, at least two of these channels (40 inputs) will be used for automatic calibration of the 20 amplifiers for gain and zero offset. By placing known signals on each amplifier, the operation of the system can be checked.

(b) The 20 variable-gain, guarded-differential input amplifiers have high-speed solid-state switches on the outputs. The gains of the amplifiers can be set manually (by switch) to various ranges from 1 to 1000. The input impedance and allowable source impedance will be sufficiently high to allow direct input of thermocouple and other low-voltage sources along with the ordinary high-voltage signals. In order to reduce noise and ground problems, the amplifiers will be able to operate at 250 V of common-mode voltage.

(c) The 20 high-speed switches on the amplifier outputs and the switch controls make up the high-speed multiplexer which feeds the analog signals sequentially to a sample-and-hold amplifier.

(d) The sample-and-hold amplifier maintains a fixed voltage input to the analog-to-digital converter (ADC) during the conversion. This amplifier has a gain of one and a maximum aperture time of one microsecond.

(e) The ADC produces a signed binary representation of the magnitude of the analog signal and presents this information to the computer. The signal voltage input to the ADC is ± 10 V and the output will consist of 14 binary bits of information.

The analog-to-digital signal conversion system will operate such that before the operation of the high-speed multiplexer is initiated, there will be a 5- or 10-ms delay after the selection of a channel of the low-speed multiplexer to allow time for the settling of the amplifiers and switching system. The high-speed multiplexer and ADC system will then reduce the 20 signals connected to the 20 amplifiers in approximately 400 μ sec. The overall system accuracy from input signal to binary representation will be $\pm 0.1\%$ while operating the low-speed multiplexer at the 10-ms delay.

(ii) The general-purpose digital computer which will perform all the arithmetic and logical control functions, as well as coordinate controller activities, will have a 24 binary bit plus parity word length and a

memory read-write cycle time of 2 μ sec or less. The initial memory capacity will be approximately 16,000 words, expandable to 32,000 words, all directly addressable. The storage elements will be magnetic cores.

The computer will contain 3 index registers as well as the usual number of arithmetic registers. The console will be capable of being located up to 20 ft from the computer main frame and will contain various displays and controls of use to the operators. These displays include the status of the interrupts, traps, flags, power, external device operations, and mode of computer operation.

(iii) One real-time and two elapsed-time clocks will be present in the system. The real-time clock will be a 23-bit binary clock operating at a frequency of 100 kc and reset on a 60-sec interval. Time increments in minutes will be stored in binary-in-computer memory and incremented each minute upon reset of the clock. This clock may be read on command or as the first word of an ADC conversion series.

The two elapsed-time clocks will be similar in construction, but will differ in frequency of counting and purpose. Both clocks must be computer programmable in that the values that are placed in the clock can be program controlled; further, on command, automatic reset on completion of count will be possible. Both may, under computer control, cause an interruption of the running program.

The differences in the two clocks will be that one operates at one megacycle and can be used to initiate a repeat high-speed sampling cycle in the analog-to-digital conversion system, and the other operates at 100 kc and can be read by the computer while it is in operation.

These three clocks will be used for timing and coordination of system and program functions.

(iv) The digital-to-analog converter channels, four in number, will be used during the simulation phase in the primary simulation of the variable transport delay times associated with the flow of coolant through interbuilding piping, and during actual plant control, they will be used to produce meter readings or display various plant parameters. The values in the "hold registers" may be changed without having to clear the register.

(v) All control of the plant, will be through on-off type of controls. Upon loss of digital controller operation, these will cause the plant parameters to fail stationary as is the case with most analog design criteria. The on-off control capability will be exercised by up to 16 level lines which will be furnished with the system. These level lines may also be used to control indicators on the main control panel. The sensing of the condition of relay contacts or interlocks can be performed by up to 32 sense lines which will be furnished with the system.

(vi) The main communication devices between the operator and the digital controller are the paper-tape reader and punch together with the typewriter. The paper-tape equipment will be used to prepare and insert programs and will also be used as a high-speed data output. The typewriter communication link will be the main source of immediate information to the operators. All requests for plant parameters and all changes in limits or demand settings will be initiated through the typewriter communication channel. In addition, other information may be typed automatically by the controller to alert the operators to changing conditions.

d. Tank Immersion Heaters. All six heater thimbles have been filled with sodium. Twenty-two operable heater cartridges are installed in these thimbles.

The heater thimble which had been slightly damaged in the air-lock explosion was successfully repaired. After filling of the thimbles with sodium, each heater cartridge was operated at full voltage (480 V a.c.) for a short period. Following this, electrical tests were performed to verify heater-element continuity and dielectric integrity of the mineral insulation. The thimbles are ready for installation in the primary tank.

4. System and Components

a. Electrical Heaters for the Rotating Plug Seal Troughs. The dipping heaters originally installed were clad with austenitic stainless steel, Type 304. Because of its high nickel content, this alloy is susceptible to corrosion by the tin-bismuth eutectic which is used as the sealant at the temperatures of the heater cladding. Heaters clad with ferritic stainless steel, Type 405, have been procured and installed to replace the original heaters.

b. Rotating Plug Seal Troughs. During the month, the seals were melted and rotated a number of times. No significant change in the operation of the rotating plug was apparent.

c. Bare Pin Control Rod. This control rod, which had been immersed in the primary sodium (storage basket) since its use last May for testing the performance of the fission product monitors (see Progress Report for May 1965, ANL-7046, pp. 2-4), was washed and dismantled in the Fuel Cycle Facility (FCF) to permit examination of the unclad fuel pin. Visual examination showed the surface condition compared favorably with that of a newly fabricated pin. Thus it appears that the uranium-fissium alloy is not adversely affected by the primary sodium. A second bare-pin subassembly will be available for fission product-monitor studies during the next reactor operating period.

Fig. 1. Calculated Radial Power Distribution in EBR-II Reactor

5. Reactor Physics

At the start of Run No. 14, the reactivity deficiency due to power operation was determined to be 71 lh, and at shutdown a value of 79 lh was obtained. The loss of reactivity due to burnup was measured during operation as 0.14 lh/MWd. This is the same value obtained during previous operation of the reactor, and includes both fuel burnup and production of Pu^{239} . The distribution of power in the reactor system as assembled for the wet critical was calculated by means of the MACH-1 code and is in very good agreement (see Fig. 1) with previous calculations as listed in the Hazards Summary Report, ANL-5719 (Addendum). This agreement gives added confidence in the ability to predict the nuclear parameters of cores with increased size and different fuel enrichment.

6. Experimental Irradiations

Exposure of the Metallurgy Division subassembly XA01, containing U-Pu-Fs alloy fuel pins, was terminated at the request of the sponsor after completion of 3940 MWd. The status of the irradiations following the completion of Run No. 14 follows:

Subassembly	Date Loaded	Accumulated Exposure (MWd)	Goal Exposure (MWd)	Experimenter	Grid Position	Subassembly	Date Loaded	Accumulated Exposure (MWd)	Goal Exposure (MWd)	Experimenter	Grid Position
XA01	5/6/65	3940	14,000	ANL(MET)	6-D-2	C220X	6/29/65	3069	3,149*	ANL(CEN)	1-A-1
XG02	7/16/65	3069	13,650	GE	7-A-1	C221X	7/24/65	3069	3,249*	ANL(CEN)	3-B-2
XG03	7/16/65	3069	19,450	GE	7-D-1	C222X	9/28/65	2248	4,328*	ANL(CEN)	5-B-2
XG04	7/16/65	3069	39,000	GE	7-B-1						
XG05	9/3/65	2642	10,300	GE-ANL(MET)	4-C-2	U-1548X	5/6/65	3940	6,080	ANL(CEN)	9-B-5
XG06	9/3/65	2642	20,600	GE-ANL(MET)	4-E-2	U-1549X	5/6/65	3940	6,080	ANL(CEN)	11-B-6
XG07	10/27/65	1905	18,600	ANL(MET)	4-D-3	U-1550X	5/6/65	3940	9,120	ANL(CEN)	13-B-7
XA08	12/13/65	820	19,800	ANL(MET)	4-F-2	U-1551X	5/6/65	3940	9,120	ANL(CEN)	15-B-8

*Minimum acceptable exposure is one-half this value.

Sixteen mixed Pu-U oxide and three mixed carbide capsules have been received from General Electric and United Nuclear Corp., respectively, for inspection and assembly into experimental subassemblies.

7. Fuel Cycle Facility

The pin-processing equipment has been modified to produce the shorter (13.5-in.) Mark-I-A fuel. The acceptance rate of the Mark-I-A fuel by the pin-processor station is over 90% (as compared to typical values of 70% for the longer Mark-I fuel).

Most of the defects that caused the fuel to be rejected in the past occurred near the ends of the cast pin. The new shear provides for more precise location of the fuel within the shear. This, plus the slightly shorter length of the sheared section of the Mark-I-A pin, allows greater latitude in selecting the section of the pin to be included between the shear blades. The new shear also seems to produce a more satisfactory pin end in that the rate of rejection due to "short shear" has decreased.

A fuel subassembly containing an unclad element (subassembly L-417X) was dismantled, and the bare pin which had been used in the reactor to calibrate the fuel-element-rupture-detection devices, was removed for examination. The pin had received a low burnup (0.03%) and had been stored in the fuel-storage basket of the primary sodium tank. It was removed from the tank to determine the effect of 200-day residence in the 700°F bulk sodium on the exposed uranium. Visual examination did not indicate any deleterious effect. Photographs and dimensional measurements are being obtained.

Examination of the fuel from subassembly C-162 is continuing (see Progress Report for December 1965, ANL-7132, p. 8). This is the first subassembly which contains fuel which had been reprocessed in the Fuel Cycle Facility and thus had received two cycles of irradiation in the reactor. The fuel had received 0.1-0.2 a/o burnup on the initial irradiation and 0.1% burnup on the reprocessed subassembly. Measurements of the sodium level in the canned fuel showed volume increases as a result of fuel swelling comparable to or slightly less than that produced in other (nonreprocessed) subassemblies of the same burnup. Measurements of pin swelling based on the gas pressure within the fuel can were also slightly lower than had been obtained on nonreprocessed fuel of similar burnup. However, a layer of scale was observed on each of the 10 fuel pins that have been declad. This phenomenon had not been observed previously. Diametrical measurements have been obtained on two of these pins and they show an increase in diameter of 3-10 mils in one case and of 4-12 mils in the other. Chemical analysis indicates that the composition of the scale is 78 w/o uranium and 10 w/o sodium. Preliminary measurements indicate that this scale weighs about 1/2 g per fuel pin. Metallographic examination is in progress.

Until this anomaly of a normal increase in fuel volume (as evidenced by measurements of sodium level and gas pressure) versus a large increase in fuel volume (as evidenced by scale formation) is resolved, it is planned to examine additional similarly produced fuel at something less than the presently maximum allowed burnup level.

Production is summarized in Table I.

TABLE I. Production Summary

Subassemblies Received			18
Subassemblies Dismantled			14
Subassemblies Fabricated			11
Subassemblies Transferred to Reactor			6
Pins Decanned			1214
Melt-refining Runs - 10	Irradiated Fuel - 7	Recycle Metal -	3
Average Pouring Yield	92.4%	93.5%	
Injection-casting Runs			8
Pin Processing (1220)			
Acceptable Pins (382 additional Chicago Mark I-A)			838
Reject Pins			59
Pins Welded			1262
Leak Testing (1296)			
Accepts			1256
Rejects			40
Bond Testing (completed runs)			
Accepts			725
Rejects			101
Surveillance - L-411, C-101			

B. General Fast Reactor Physics

1. ZPR-3

Following is a detailed description of Assembly 47, a mockup of SEFOR (see Progress Report for November 1965, ANL-7122, pp. 19-21) at reactor loading No. 15, the loading at which the assembly was first critical.

a. Initial SEFOR Criticality. An interface view of Half No. 1 of the reactor is shown in Fig. 2. All of the drawer type numbers are shown

in their proper locations. Half No. 2 of the assembly is loaded as the mirror image of Half No. 1, except for the location of the C-6 and C-16 type drawers in the core. These drawers will be discussed later.

Fig. 2. Interface View of Half No. 1 of Assembly 47, ZPR-3, for Loading No. 15

Two types of plutonium fuel are used in the mockup. The original or "old" fuel on hand was plutonium with 1.1 w/o aluminum jacketed in stainless steel, in the form of plates of 1/8-in. nominal thickness. The additional plutonium required for this assembly was obtained as a Pu-U-Mo alloy, called "new." The plutonium enrichment is 20%. It is also canned in stainless steel and is 1/4 in. thick. The new plutonium was loaded in core region 1A and the old plutonium in core region 1B (see Fig. 3) to provide a negative overall temperature coefficient of reactivity for this assembly.

Fig. 3. Horizontal View of Assembly 47 of ZPR-3

Table II lists the actual material composition of the regions in the assembly with all control drawers inserted. With this configuration, the measured multiplication constant, k , was 1.00030 ± 0.00003 . The mass of this loading included 314.31 kg $\text{Pu}^{239+241}$ in a core volume of 527.6 liters.

TABLE II. Composition of Assembly 47 for Loading No. 15

Reactor Region			Material Composition (g/cc)											
Name	No.	Volume (L)	Pu ²³⁹ + Pu ²⁴¹	Pu ²⁴⁰	U ²³⁸	U ²³⁵	Be	O	Mo	C	Na	Al	304SS	Ni
Core (New Pu)	1A	203.1	0.5875	0.0525	3.06	0.006	0.085	0.416	0.082	0.067	0.253	0.313	1.31	
Core (Old Pu)	1B	324.5	0.6007	0.0287	3.12	0.006	0.081	0.406		0.067	0.252	0.346	1.43	
Radial Na / SS	2	661.5									0.235	0.022	2.16	
Radial Reflector	3(Ni)	579.2										0.276	0.60	6.36
Radial Reflector	3(SS)	155.6										0.274	6.29	
Axial Reflector	4	195.5								0.035		0.239	1.62	4.25
Axial Na / SS	5	127.7								0.075		0.482	3.58	

b. Fuel Expansion Experiment. A uniform expansion of all the fuel columns, including the depleted uranium, was made. This simulated the response of the reactor to an excursion before the heat generated in the

fuel has been transferred to the coolant and the structural material in the core. The expansion was made by removing two of the four $1/8 \times 1/4 \times 2$ -in. pieces of 100% aluminum from the axial reflector at the end of each fuel column in the core drawers and placing the spacers between fuel pieces. The fuel columns then extended into the axial reflector. Table III gives the location of these two spacers in each fuel column in each type of drawer loading. All fuel columns were expanded except those in the 10 control drawers. Expansion of the fuel columns in Half No. 1 resulted in loss of 242 lh. The expansion in Half No. 2 resulted in a loss of 203 lh for a total loss of 445 lh for the complete expansion of the fuel columns.

TABLE III. Location of Spacers for
Expanded Fuel Experiment

Drawer Type	Spacer Location (Distance from Interface, in.)	
	Half No. 1	Half No. 2
C-1	4 and 7	5 and 11
C-2	4 and 7	5 and 11
C-3	4 and 7	5 and 11
C-4	4 and 7	5 and 11
C-5	4 and 7	5 and 11
C-6	4 and 7	5 and 11
C-11	3 and 10	3 and 11
C-12	3 and 10	3 and 11
C-13	3 and 7	3 and 11
C-14	3 and 7	3 and 11
C-15	3 and 7	3 and 11
C-16	3 and 7	3 and 11

c. Fission and Reactivity Distribution. To determine the power distribution in the SEFOR reactor, axial fission traverses were made with U^{235} , U^{238} , and Pu^{239} fission counters and a traverse with a BF_3 proportional counter. The traverse tube was placed in the center of the 1-P-16 and 2-P-16 drawers. This placed the traverses 2 in. from the axial centerline of the core. The curves are shown in Figs 4 through 7. Radial traverses will be run at a later date.

The General Electric Co. is planning to install a fast reactivity excursion device (FRED) through the center of the SEFOR reactor core. Rapid changes in reactivity will be made by the movement of a B_4C slug. To help evaluate the effectiveness of this device, a small sample containing 3.25 g of B^{10} was moved axially through the assembly by the traverse drive mechanism, and the worth of B^{10} as a function of position in the assembly was measured. Figure 8 gives the results of the B^{10} worth corrected for the worth of the steel container and the drive rod.

Fig. 4. Axial Traverse with U^{235} Fission Counter in Assembly 47, ZPR-3

Fig. 5. Axial Traverse with U^{238} Fission Counter in Assembly 47, ZPR-3

Fig. 6. Axial Traverse with Pu^{239} Fission Counter in Assembly 47, ZPR-3

Fig. 7. Axial Traverse with BF_3 Counter in Assembly 47, ZPR-3

Fig. 8. Axial Worth of 3.25 g Boron-10 in Assembly 47, ZPR-3

2. ZPR-6

a. Reactivity Worths in Assembly 4Z. The reactivity associated with each of several samples was measured at $r = 16.5$ cm and $z = 0$ in Assembly No. 4Z (see Progress Report for May 1965, ANL-7046, p. 14). The symbols r and z refer to the radial and axial dimensions, respectively. Previously, this type of measurement was performed at the center of the core (see Progress Report for October 1965, ANL-7115, p. 28).

The results of the measurements and the values calculated from the use of two cross-section sets are given in Table IV. Cross-section

TABLE IV. Comparison of Measured and Calculated Worths of Various Materials at the Center and at 16.5 cm from the Center Radially in Assembly No. 4Z of ZPR-6^a

Sample (Weight, in g)	Net Measured Worth per Sample (1h)		Worth Calculated with Set 224 (1h)		Worth Calculated with Set 801 (1h)		$(Z_0)_{\text{measured}}^a$	$(Z_0)_{224 \text{ Set}}^a$	$(Z_{16.5 \text{ cm}})_{\text{measured}}^a$	$(Z_0)_{224 \text{ Set}}^a$
	At Center	at 16.5 cm.	At Center	At 16.5 cm	At Center	At 16.5 cm				
U ²³⁵ (13.869)	+0.611 ± 0.020	+0.568 ± 0.030	+0.664	+0.627	-0.760	+0.732	1.000	1.000	1.000	
U ²³⁸ (1153.8)	-3.857 ± 0.010	-4.212 ± 0.014 ^b	-4.018	-3.77	-4.470	-4.140	-6.31 ± 0.032	-6.051	-7.415 ± 0.052	1.043
Pu ²³⁹ (121.55)	+8.446 ± 0.200	-	+7.988	-	-	-	+13.82 ± 0.038	+12.030	-	1.149
U ²³³ (146.71)	+11.643 ± 0.200	-	+12.831	-	-	-	+19.055 ± 0.037	+19.323	-	0.986
Na (51.38)	-0.049 ± 0.005	-0.032 ± 0.007	-0.092	-0.0788	-0.636	-0.0542	-0.080 ± 0.105	-0.138	-0.056 ± 0.230	0.580
Fe (488.0)	-0.739 ± 0.007	-0.663 ± 0.014	-0.954	-0.886	-1.082	-0.987	-1.209 ± 0.032	-1.437	-1.167 ± 0.055	0.841
Ni (546.0)	-1.148 ± 0.010	-1.013 ± 0.004	-1.650	-1.540	-1.717	-1.550	-1.879 ± 0.033	-2.485	-1.783 ± 0.052	0.756
SS 304 (499.6)	-0.849 ± 0.010	-0.767 ± 0.020	-1.224	-1.073	-	-	-1.389 ± 0.033	-1.843	-1.350 ± 0.055	0.754
C (103.0)	+0.194 ± 0.010	+0.181 ± 0.051	-0.154	-0.140	+0.0718	+0.080	+0.317 ± 0.060	-0.232	+0.318 ± 0.220	1.366
Be (114.31)	+1.107 ± 0.010	+1.098 ± 0.040	+0.352	+0.324	-	-	+1.811 ± 0.033	+0.530	+1.933 ± 0.063	3.417
Al (166.90)	-0.297 ± 0.013	-0.257 ± 0.020	-0.469	-0.435	-0.420	-0.377	-0.486 ± 0.055	-0.706	-0.452 ± 0.083	0.688
B ¹⁰ (33.30)	-23.339 ± 0.200	-20.60 ± 0.20	-30.363	-28.498	-	-	-38.198 ± 0.033	-45.727	-36.267 ± 0.052	0.835
(CH ₂) _n (59.15)	+19.751 ± 0.010	+17.78 ± 0.10	+9.879	+9.405	-	-	+32.325 ± 0.033	+14.878	-31.302 ± 0.052	2.173
W (1108.0)	-5.742 ± 0.010	-5.268 ± 0.004	-6.441	-6.040	-	-	-9.397 ± 0.033	-10.541	-9.274 ± 0.052	0.891
Mo (599.0)	-3.415 ± 0.010	-3.062 ± 0.020	-3.797	-3.527	-	-	-5.589 ± 0.033	-5.143	-5.390 ± 0.053	0.990
Nb (481.2)	-3.791 ± 0.010	-3.543 ± 0.022	-4.742	-4.441	-	-	-6.204 ± 0.033	-7.141	-6.238 ± 0.052	0.869
Ta (924.70)	-11.907 ± 0.200	-9.98 ± 0.20	-13.890	-13.037	-	-	-19.487 ± 0.033	-20.918	-17.570 ± 0.052	0.931
B ₄ C (59.62)	-6.410 ± 0.025	-11.50 ± 0.20	-7.829	-7.342	-	-	-10.490 ± 0.033	-11.790	-20.246 ± 0.052	0.890
Zr (406.0)	-0.650 ± 0.005	-0.594 ± 0.002	-0.614	-0.566	-	-	-1.064 ± 0.033	-0.925	-1.046 ± 0.052	1.150

^aCalculations were done with MACH 1, a one-dimensional diffusion code.

^aZ is defined as the worth of a material to that of U²³⁵. The subscripts 0 and 16.5 refer to positions $r = 0, z = 0$, and $r = 16.5$ cm and $z = 0$, respectively.

^bThis sample was different from the one used at the center.

Set 224 uses 22 groups and contains many nuclides whose cross sections have been ELMOE averaged for typically large carbide cores. Cross-section Set 801 uses 25 groups and has been ELMOE averaged specifically for Assembly No. 4Z. The neutron spectra determined by both sets are about the same. In Table IV, the column designated $(Z_0)_{\text{measured}}$ is the ratio of the worth of a material to the worth of U^{235} at the center of the core. The same ratio measured at 16.5 cm from the center radially is designated by $(Z_{16.5})_{\text{measured}}$. Comparison of these two columns confirms that the neutron spectrum at these two positions is essentially the same. The differences in the values of Z at the two positions are within the statistical accuracy for all of the samples except for U^{238} and B_4C .

In the case of the U^{238} , two different samples were inadvertently used at the two positions. This may well be the source of error. In the case of the B_4C sample, the reason for the discrepancy is still unknown.

The last column of Table IV reflects the accuracy of the calculations. The values in this column were derived from the measured and calculated numerator parts of the perturbation expression. Thus, these values depend on the real and adjoint fluxes in the volume that the sample occupies and on the cross section of the sample. If the cross section is accurately known and if the calculated neutron spectrum is correct, then the value for that material should be unity. The magnitude of the deviation from unity reflects the magnitude of the errors in the cross sections and in the neutron spectrum. The cross-section sets used in this analysis do not seem to do justice to the worth of materials of low atomic number.

The calculated values are larger than the measured values except for C, Be, and polyethylene. The worths of U^{235} and U^{238} predicted with the 224 Set are in better agreement with experiment than are the values obtained from the 801 Set.

The effect of the thickness of the U^{235} sample on its worth was also tested at $r = 16.5$ cm and $z = 0$. The results are shown in Table V.

TABLE V. Worth of U^{235} as a Function of Thickness

Sample	Weight (g)	Net Worth (Ih)	Worth/kg (Ih)
3(2 x 2 x 0.005 in.)	13.869	$+0.568 \pm 0.030$	40.95 ± 2.16
6(2 x 2 x 0.005 in.)	27.719	$+1.164 \pm 0.030$	41.99 ± 1.08
12(2 x 2 x 0.005 in.)	55.029	$+2.367 \pm 0.040$	43.03 ± 0.73

b. Fission Traverses in Assembly No. 4Z. Fission rates in U^{235} and U^{238} foils were mapped throughout Assembly No. 4Z. Radial traverses were taken 7 cm from the interface of the halves in order to eliminate the influence of the drawer front walls and of the residual gap; axial traverses on the

core axis, at the edge of the central zone, and in the driver. The fine structure of the U^{238} fission rate was measured across a sequence of three drawers in the core center. In order to study the influence of the zoning on the flux distribution, most foils were placed at the outer edge of the core and in the buffer and driver regions. The influence of the orientation of the material plates and of the irregular core shape was studied by making radial traverses in horizontal, vertical, and diagonal directions from the center of the core.

All foils were placed in vertical orientation between adjacent material plates. They were irradiated for 20 min at a reactor power of about 150 W. The activation could be determined with an accuracy of about 0.5% for the U^{235} foils and of 2.5% for the U^{238} foils. Figures 9-13 show the results of the measurements. In the radial traverses of the U^{238} activation (see Fig 10), the average over the measured fine structure (see Fig. 13) was used for drawers in the central zone. In the outer regions, where no fine-structure measurements were available, only the results for U^{238} foils without direct contact with enriched fuel plates are plotted.

Fig. 9. Radial Traverse of U^{235} Fission Rates in ZPR-6 Assembly No. 4Z

Fig. 10. Radial Traverse of U^{238} Fission Rates in ZPR-6 Assembly No. 4Z

Fig. 11. Axial Traverse of U^{235} Fission Rates in ZPR-6 Assembly No. 4Z

Fig. 12. Axial Traverse of U^{238} Fission Rates in ZPR-6 Assembly No. 4Z

Fig. 13. U^{238} Fine Structure through a Three-drawer Sequence in the Central Zone in ZPR-6 Assembly No. 4Z

The measured values were compared with diffusion and transport calculations for the radial traverses and with a cosine distribution for the axial traverses. The transport calculations agree quite well with the measurements, whereas diffusion theory yields considerable deviations and does not show the full extent of the fluctuations of the U^{238} fission rate between central zone, buffer, and driver.

The direction in which the radial traverses were taken had little influence on the measured results. All axial traverses (with the exception of U^{238} values very near to the blanket) very closely follow a cosine distribution and yield a reflector savings of 16.6 cm when extrapolated to zero flux.

3. ZPR-9

a. Assembly No. 9. Studies of Assembly No. 9 have been completed. Spatial-dependent reaction rates were determined for U^{233} , U^{235} , U^{236} , U^{238} , Pu^{239} , Pu^{237} , and for a $B^{10}F_3$ counter. Fission reactions were determined from thin coatings of the various materials on a solid-state detector. Reaction rates were mapped out through the core midplane in a radial direction. Detectors were driven through a 3/4-in. voided tube starting at the outer edge of the radial reflector, going through the core region and ending on the opposite outer reflector edge. During these measurements, the reactor flux level was held constant by on-line computer control. The data, read directly on punched tape, have yet to be analyzed.

The spatial dependence of the reactivity worths of a number of small samples were also determined. In these measurements the effect of the void tube on the reactivity measurements was determined by pairs of measurements. In the first series of each pair, the samples were sandwiched between long lead rods to eliminate streaming through the void tube. In the second set, the reactivity effects of the same samples were determined in the absence of the lead rods. Samples of U^{235} , B^{10} , polyethylene, borated polyethylene, rhenium, tungsten, dysprosium, gadolinium, and europium were measured in this manner.

In these measurements, the probe drive unit was used to move the samples through the core and reflector in the same void tube that was used to determine the reaction rates. As the samples were driven at a constant but slow speed, the computer determined a least-squares-fitted period every tenth of a second. Reactivities were then determined from computer solution of the inhour equation. Probe drive speeds were adjusted so that the change in worth every tenth of a second was less than the error propagated through the inhour equation from the period fits. Measurements were made for each sample driven in both directions through the core as a test of the reliability of the computer program to solve both positive and negative reactivity changes. Excellent agreement between measurements was observed for all cases. Preliminary analysis of the data does not

indicate that there is significant amount of streaming caused by the void tube through the core. As was the case with the reaction rate measurements, these data are on punch tape and are currently being analyzed.

One of the routine measurements made in the center of all of the ZPR-9 assemblies is the $U^{238}:U^{235}$ fission ratio. This measurement is normally made by putting a pair of foils (a U^{235} and a U^{238} foil) at the core midplane. A second pair in the same region but oriented 90° from the first pair is irradiated simultaneously. After the irradiation the fission product Mo^{99} is chemically separated from each foil and counted to determine the number of fissions in each sample. These results gave a measured $U^{238}:U^{235}$ fission ratio of 0.04307 for both foil pair orientations.

The central worth of a 142.53 g sample of U^{233} was determined by the calibrated-rod technique. The results gave a central worth of 359.2 lh/kg of U^{233} .

A series of measurements determined the approximate worths of various sections of aluminum oxide reflector. In these measurements, varying amounts of reflector material were removed from different regions of the reflector and the worth of the Al_2O_3 removed was determined relative to a void with calibrated control rods. In the first series of measurements, the first 2 in. of radial reflector surrounding the movable half of the core was removed from an approximate 56° solid angle. The resulting loss of reactivity amounted to 204.3 lh. Extrapolating this loss to 360° solid angle and to the full core length indicated that the first 2 in. of radial reflector was worth approximately 6.18% $\Delta k/k$. In the next series, the worth of the bottom 2 in. of the radial reflector was determined for a set of disc-shaped voids, each 2 in. high and of increasing radii until the bottom 2 in. of radial reflector (from the core-reflector interface to the outer radius of the reflector) had been removed. Measurements were made only through a 90° solid angle and had to be extrapolated to a 360° sector. In the first 5 cm of the radial reflector (in the bottom 2 in. of the core), the reflector material had a measured worth of 8.15 lh/kg of aluminum oxide relative to a void. Extrapolation of the measured data indicated that the total worth of the bottom 2 in. of the radial reflector was 1.4% $\Delta k/k$. Similar measurements and extrapolations for 4-in.-high coin-shaped voids at the bottom 4 in. of the reflector removed a total of 2.7% $\Delta k/k$. Finally, the same measurements repeated at the core midplane indicated that the first 5 cm of the reflector had a worth of 16.8 lh/kg of aluminum oxide relative to a void. Extrapolating these measurements over a 360° sector gave the indicated worth of a 2-in.-thick disc of the radial reflector at the core midplane of 2.5% $\Delta k/k$.

The final measurements in Assembly No. 9 consisted of a determination of the hydrogen void coefficient in the core. In these measurements hydrogen in the form of polyethylene sheets, 20 mils thick, were added

to the bottoms of each of the core drawers. This amount of polyethylene corresponds to a hydrogen density of 0.959 g of hydrogen per liter of the core. The measured worth of polyethylene at the core axis was 3.02 lh/g of hydrogen and at the core edge was 1.66 lh/g of hydrogen. After all drawers had polyethylene loaded into them, there were 120.4 g of hydrogen in the core and this amount of hydrogen added 0.47% $\Delta k/k$.

b. Assembly No. 8. After Assembly No. 9 was dismantled, Assembly No. 8 was built into the ZPR-9 facility.

Assembly No. 8 in ZPR-9 was designed to study the properties of a beryllium oxide-reflected tungsten-based core. The core represents a mockup having 40 v/o $U^{235}O_2$ fuel, 33 v/o tungsten diluent, and the equivalent of about 25 v/o void. The actual loading has the same core composition as Assembly No. 9, which utilized metallic uranium and Al_2O_3 to simulate oxide fuel and void. The radial reflector consists of 10.6 cm of high-density BeO. The radial reflector was surrounded with a 46.4-cm-thick aluminum isolation blanket to prevent flux distortions caused by backscattering from the matrix, the table bed, and the floor of the assembly chamber. The axial reflector is 43 cm of aluminum. A material producing higher albedo would have been preferred, but aluminum was chosen to insure a negative coefficient of thermal expansions in the axial direction, since it conforms to the specified requirements for safe operation.

Calculations, by diffusion theory with a sequence of one-dimensional slab and cylindrical calculations that represented the two-dimensional system, indicated a critical mass of 283 kg of U^{235} . The Argonne modified Hansen-Roach cross-section set was used.

The core was loaded to a length of 51 cm. Criticality was achieved with a fuel inventory of 283.9 kg of U^{235} , a core-equivalent cylindrical radius of 31.74 cm, and a volume of 161.4 liters. The mass is corrected only for the excess reactivity available in the control rods. No correction has been made for the heterogeneity of the fuel, the irregularity of the actual radial surface of the core, or the gap between the assembly halves.

Beryllium oxide at the outer edge of the reflector had a measured worth of 6.46 lh/kg of beryllium oxide, and fuel at the outer edge of the core had a measured worth of 25.4 lh/kg of U^{235} . Further studies will include measurement of the spatially dependent reaction rates and reactivity traverses, as well as determination of the reactivity coefficient for the hydrogen addition to the core.

4. ZPPR

A supplement to the ZPPR Preliminary Safety Analysis Report is in final preparation. A backup containment structure has been added to the reactor containment roof, which increases the ability to attenuate the products of either a credible or design-basis reactor accident.

The following is the current status of the reactor components:

a. Bed and Tables. Giddings and Lewis Machine Tool Company, Fond du Lac, Wisconsin, are within two weeks of their originally estimated time schedule, prepared in August 1965. Shipping date for the bed and tables is scheduled for late May 1966. Since approval for construction has been delayed, it will be necessary to provide storage for the completed bed and tables if the vendor ships the units as contracted.

b. Matrix Drawers. The vendor has shipped two sample drawers for inspection to ANL-Idaho. Approval to proceed with production will be given only if the two prototype drawers meet the requirements for ZPPR.

c. Matrix Tubes. The first batch of 600 tubes has been completed and inspected by the vendor. Of the second batch, 150 tubes have been fabricated, but the drawer notches have not been punched. ANL will inspect the first two batches (1200) when they are completed.

The tubes are presently being stored at the vendor's plant. ANL cannot proceed with the shipping and bundling of the tubes because AEC funds have not been released. Previous experience on tube bundling indicates that this is one of the long lead-time items which should be expedited to prevent delays in the final reactor assembly.

d. Fuel-rod Drives. The vendor, McGee and Hogan Machine Works, has completed and shipped nine rod drives. An ANL representative inspected the first three rod drives and found them to perform satisfactorily. Two minor items were to be corrected: (a) a screw which holds an electrical cable interferes with the forward mechanical stop, and (b) the housing around the magnet requires a clearance slot for the electrical cable. Damage to a guide rod during fabrication is delaying the completion of the 10th rod drive. A new guide rod is being manufactured.

ANL will make a final inspection of the rod drives after they have been delivered.

e. Nuclear Instrumentation. The nuclear instrumentation is 33% complete. Design and development work is essentially completed. All chassis are in production status. A transistorized fail-safe trip circuit is being thoroughly checked out for proper operation.

f. Gamma-monitoring Instrumentation. This instrumentation is the same as for EBR-II to provide an integrated monitoring system. The gamma-monitoring instrumentation is 25% complete.

All other reactor items are delayed due to withholding of procurement funds.

C. General Fast Reactor Fuel Development

1. Zero-power Reactors

a. ZPPR Fuel Elements. A commercially low-cost method for making ZPPR fuel elements is under development. The objectives are as follows:

- (i) to design and develop U-Pu-alloy plate fuel elements that provide prompt transmission of thermal expansion and safe containment of the fuel;
- (ii) to investigate potentially low-cost methods for fabricating core plates of uniform plutonium content and precise dimensions;
- (iii) to develop methods for forming the jacket components;
- (iv) to demonstrate methods for jacketing and seal welding the core plates in the jacket components that minimize contamination problems;
- (v) to test methods of pilot production of ZPPR fuel elements and to develop inspection methods and standards;
- (vi) to draft technical specifications for the procurement of ZPPR fuel elements.

The reference alloy for fabrication development is U-25 w/o Pu-2.5 w/o Mo. Alternative alloys are also being investigated that may be more desirable for ZPPR fuel elements.

Three potential methods for fabricating the ZPPR alloy have been considered: (1) precision casting, (2) casting and rolling, and (3) casting and extrusion. Time limitations dictate the early selection and development of the most promising of these methods. The reference alloy is hard and non-ductile. Very little plasticity develops until it is heated to above 600°C. Because of the high hardness of the material at room temperature, carbide tools have been required for machining. Further development of machining methods is being undertaken for cutoff, finishing, and weight adjustment.

Emphasis in the pilot production of jacket components has been placed upon techniques that yield good precision and freedom from jacket defects. Jacket-component fabrication must be adaptable to large-scale production and minimum tooling cost.

Contamination control during fuel-element assembly is a very real and a potentially costly problem. An alpha-contamination level of

400 DPM on the surface of the fuel elements represents 0.000,000,001,4 g Pu. Techniques are being developed to minimize contamination of the jackets during assembly. Weld contamination has been a particular problem because of the difficulties in decontaminating welds.

Welding and weld inspection have caused serious difficulties in the commercial procurement of fuel elements. This is partly because few vendors realize the precision of welding fixtures and arc control required to weld thin metals satisfactorily. It is partly due to the difficulty in setting up inspection standards and in describing unsatisfactory welds. The welding phase of this program will attempt a description of the parameters required to make satisfactory welds in the fuel elements and to provide inspection standards.

ALL DIMENSIONS IN MILLIMETERS

NOMINAL	25	51	76	102	127	152	178	203	229
CORE LENGTH	20.42	45.82	71.22	96.55	121.9	147.3	172.7	198.1	223.5
JACKET LENGTH	20.32	45.72	71.12	96.44	121.8	147.2	172.6	198.0	223.4
JACKET LENGTH	25.65	51.05	76.45	101.8	127.3	152.7	178.1	203.5	228.8
JACKET LENGTH	25.40	50.80	76.20	101.5	126.9	152.2	177.6	203.0	228.3

Fig. 14. ZPPR Reference Element

design will allow cutting cores from cast or fabricated U-25 w/o Pu-2.5 w/o Mo alloy bars. The jackets may be made from precision-formed rectangular

(i) Design of Fuel Element.

The reference design for the ZPPR fuel element is shown in Figs. 14 and 15. Although most of the fuel elements will have the cross section shown in Fig. 14, a small number will be required that are one-half the thickness of the plates shown. Nine different lengths of fuel elements are required, each having the same cross section and the same end closures. This de-

Fig. 15
Components and Proto-
type ZPPR Fuel Element

tubing and the end plugs cut from rectangular wire. It is believed that this design will reduce the tooling cost when compared to methods that require separate molds or dies for each size of fuel element. Cross-sectional clearance is provided so that the plutonium alloy core plates can be loaded into the jackets through a contamination-preventing funnel and sheath.

It is intended that one end plug be welded in place and the joint leak detected before core loading. The core will be loaded and the opposite end plug inserted. The assembly will then be clamped in a welding fixture that squeezes the core between the two end plugs while the final weld is being made. This weld must be made under a partial vacuum so that the jacket does not inflate at Idaho altitude pressure and at the anticipated fuel-element temperatures. Contact between the core and jacketing provides prompt transmission of thermal expansion to the jacket ends. The end plugs are sufficiently thick that they can be machined parallel if welding causes distortion.

(ii) Casting. Alloying and precision casting methods have been well-developed for the U-Pu-Mo alloys.¹ The properties of these alloys were described by Kelman *et al.*² The relatively low rate of atmospheric oxidation and low pyrophoricity of these alloys at room temperature, their high compressive strengths, and their high coefficients of thermal expansion were important factors in the selection of U-Pu-Mo alloy. The alloy cannot be cold straightened or otherwise room-temperature deformed because of its low ductility.

The casting properties of the alloy are good. Preliminary tests show that the casting properties of the U-Pu-2.5 w/o alloys are similar over the range from 20 w/o to 30 w/o Pu. Only slight modification of the casting methods developed for 178-mm-long ZPR-3 SEFOR critical test elements is required to produce 25.4-cm long, precision-cast bar sections. Longer cast bars may also be practical, but it was believed that these would have very little advantage over multiple cast 254-mm lengths.

Difficulties were experienced by the ZPR-3 SEFOR contractor with carbon pickup from ceramic-washed carbon crucibles.³ Ceramic crucibles can be used for melting the alloy but with increased dross losses. Large ceramic crucibles have a poorer thermal-shock resistance than carbon crucibles and must be used more carefully.

¹Shuck, A. B., Pilarski, E. M., Hins, A. G., and Burton, G., Jr., Development of a Process for Manufacturing ZPR-III U-Pu-Mo Test Fuel Elements, ANL-6955 (March 1965).

²Kelman, L. R., Rhude, H. V., Schnizlein, J. G., and Savage, H., "Metallic Plutonium Alloys for Fast Critical Experiment," Third International Conference on Plutonium, Institute of Metals (November 22-26, 1965).

³Collett, J. G., Benson, E. M., Gerrish, R. W., and Bruggeman, L. D., Zero-Power Reactor III Fuel Fabrication, Trans. ANS 1965 Winter Meeting, Vol. 8, No. 2, 354-355 (November 15-18, 1965).

Of several oxide crucibles tried, ZrO_2 seems to give the most satisfactory overall performance. The Zirconium Corporation of America has suggested the use of hydrogen refired crucibles to reduce drossing. A suboxide layer is produced on the surface of the ZrO_2 crucibles that is said to reduce the amount of oxidation of molten metals.

The cast alloy blackens in air but is not pyrophoric. Castings have presently been made to a length of 254 mm from which shorter individual core plates are being cut.

Fig. 16. ZPPR Bar Casting before Removal from Mold

One of these castings is shown in its mold in Fig. 16. The dimensions of the core plates have been chosen such that each 25 mm of length weighs approximately 100 g. Each 254-mm casting will weigh approximately one kilogram. Individual process studies must be made by each contractor to determine maximum safe-batch limits in his plant. But it appears that multiple-mold techniques can be developed that will allow casting of batches as large as 30 kg. Glovebox handling of larger batches must be mechanized.

Precision casting, either of the individual pieces or of precision-sectioned bar, appears to be a practical method for the making of the core plates. This is the present method of choice.

(iii) Secondary Working.

Secondary working requires the casting of billets, hence it does not eliminate the casting phase from the process. The costly rolling mills or extrusion presses used to fabricate

plutonium cannot be used for other products because they become radioactively contaminated. The metals and machinery must be gloveboxed, not only for personnel protection, but also for the high-purity inert atmosphere required for hot working. Because maximum batch limits can be precision cast in multiple molds with acceptably low recycle, the economic justification for secondary working may be questioned. Investigation of rolling and extrusion techniques was undertaken because several potential fabricators expressed an interest in these methods.

The brittleness of the alloy presents a problem in secondary working. Tests were made with small billets rolled on a 12.7 x 20.3-cm two-high rolling mill. The rolls were electrically heated to 260°C, the maximum temperature obtainable with the 2-kW heaters in the rolls. Rolling tests were made at 550, 610, and 650°C with standard 5.2-mm-thick, 25 x 50-mm fuel-plate castings. Severe brittle cracking occurred at small reductions at 550°C. Lateral cracking also occurred with 30% reductions at 610°C. Thirty percent reductions were accomplished successfully at a billet temperature of 650°C. The billet oxidized excessively in the glovebox atmosphere containing less than 0.015% oxygen and 60 ppm H₂O. A limited number of tests were made with molten salt coatings to determine whether a neutral salt bath might reduce oxidation. The method was found to be messy and somewhat hazardous for glovebox operation, and was discontinued. The tests indicated that hot working could be done at sufficiently high temperatures, but did not show sufficient promise to warrant tooling up one of the large mills with heated rolls for full-scale tests.

Extrusion is a method that works metal largely by compressive forces. In the extrusion tests under way, the alloy appears to have a tendency to attack ferrous dies at the near 800°C temperature required for extrusion. Protective lubricants or ceramic extrusion tools will be required if die erosion and container-sticking problems cannot be eliminated. Extrusion in protective cans is not recommended because of the imprecision of the method and the problems associated with jacket removal.

(iv) Machining. Considerable machining experience was gained with the U-20 w/o Pu-2.5 w/o Mo alloy. Tests of the U-25 w/o Pu-2.5 w/o Mo alloy have revealed similar machining characteristics. High-speed steel tools, used dry at room temperature, did not stand up. Carbide tools performed reasonably well. Although solid carbide cutoff saws maintain their cutting edge, they are subject to breakage and their cost is over \$50 each. A large amount of spallation and edge breakage resulted during cold machining. Chip formation was of a brittle and powdering nature.

A number of holes were drilled in the alloy fuel plates. High-speed steel drill points blunted immediately and were not successfully used at room temperature. Carbide drills were used but edge breaking occurred around the drilled holes. It was found that if the plates were heated to above 200°C they could be drilled with the high-speed drills. Plate breakage was reduced and a normal-looking drill chip resulted.

This led to the design of an electrically heated milling fixture and gang slitting saws to part the plates from the alloy bars. The fixture has just been completed and has been tested with high-speed steel cutoff saws. The hot milling setup is shown in Fig. 17. Tests were made at 100, 125, 150, 175, and 200°C with a 1.6 x 76-mm-dia slitting saw having 28 teeth.

Fig. 17. Hot Milling Setup for Multiple Sawing of Four ZPPR Bars

The speed was 80 rpm and feed was 0.6 mm/min. Improved machinability was obtained in tests above 150°C. Edge spallation was greatly reduced at 175°C and essentially eliminated at 200°C, although surface oxidation was increased. Normal cut machining chips were produced at 175 and 200°C. The machined surfaces are shown in Fig. 18.

(v) Jacket Fabrication.

Type 304-L stainless steel jackets are preferred for two reasons: the stainless steel is sufficiently tough to survive handling, and it is a normal component of most of the reactor cores to be mocked up in ZPPR. In the past, envelope-type jackets have been wrapped up and seal welded from sheet metal. It has been very difficult to maintain satisfactorily high precision by this method. Individual tooling was required for each jacket size, and time-consuming hand operations were necessary that made the jackets costly. Drawn jackets were used for the ZPR-3 SEFOR fuel but the projecting edge welds were objectionable.

Fig. 18
Hot Machined Surfaces
of ZPPR Castings

Fabrication of jacket sleeves from rectangular precision-formed tubing appears to offer a low-cost, high-production method for making jacket envelopes. The varying sizes will be cut from long lengths of the tubing. Development of the tube-forming process has been undertaken at Argonne.

The steps of tube forming are shown in Fig. 19. A 0.4-mm-wall tube is drawn to the required diameter. The 6.1-mm sides and edges are formed over a mandrel by means of dies in a press brake. The 50-mm sides are flattened in a second press brake operation. The resulting, slightly undersized, preformed tube is drawn over a stretch mandrel in a rectangular forming box to finished dimension. The resulting rectangular sections can be inspected and leak-detected before they are cut to jacket lengths. In production, end plugs will be cut from precision-drawn rectangular-section wire. Jacket-component machining should be reduced entirely to cutoff deburring, finishing, and cleaning operations.

Fig. 19. Steps for Forming Precision, Rectangular Jacket Tubes

Some sample jackets and jacket sleeves were produced during process development and were intended for experimental purposes only. Several of the jackets were sectioned and inspected metallographically. With few exceptions the jackets were found to be of good quality. The mean width and thickness was on the low side of the specified range and should be increased. The variation of dimensions over the length of the jacket was well within the specified tolerances. The butt weld that closes the jacket sleeve was sound and uniform. No weld section was found to have a thickness of less than the specified minimum of 0.31 mm. There is some convexity of the jacket walls, but this should draw down when the jackets are partially evacuated.

The chief problem appears to be a thinning of the jacket sleeves in the corners. The first jackets were formed with a 0.43-mm radius in the corners. The minimum wall thickness in the corners was less than 0.31 mm. The corner radius on subsequent jackets was increased to 0.58 mm and then to 0.66 mm. The minimum corner thickness

increased only slightly. This problem has not yet been solved, but it appears that the corner thinning does not occur in corner bending.

Very careful handling is required to keep plutonium contamination from the outside of the jackets and weld joints. A disposable loading funnel is used to prevent contamination of the jacket lip. The small end of the funnel must be thin enough to be inserted well into the jacket. In the past these funnels have been improvised from 50- to 100- μ aluminum foil, brass shim stock, or plastic film. Because of the small clearances, the hand-made funnels have to be extremely precise and were costly to make. The vacuum-formed plastic funnels lacked the required rigidity, and particles for plastic tended to be carried into the jacket as the fuel plates were loaded. Impact extrusions and draw dies are being made up for the high-rate production of aluminum loading funnels. It is believed that these will provide the necessary rigidity and precision at lower cost.

Those sample jackets not destructively tested are being made into dummy fuel elements. The jackets are slightly wider in the

center than at the edges. This convexity will undoubtedly diminish as the jacket process develops; however, some of the convexity should be removed by the negative pressure within the finished element. A group of 17 jackets is being used to determine the degree of convexity that can be removed. Their maximum thickness as received has been measured, a steel core has been loaded into each jacket, and the jackets are being welded. The dummy elements will be measured and the two sets of measurements will be compared to determine the effect of pressure.

Fig. 20. Vacuum Welder Used for ZPR-3 and ZPPR Fuel Elements

(vi) Welding. After insertion of the core plates and the end plugs, the assembled elements will be clamped in a chill fixture, shown in Fig. 20, designed to cool the four sides of the jacket and the end plugs, and to force the two end plugs against the core plate while

welding. The welding fixture will be mounted in a vacuum welder, which will allow the welds to be made under about one-half atmosphere pressure of A + He gas mixture. The welder is shown in Fig. 8.

Both TIG weld and electron-beam weld methods of closure welding are being investigated. Because we presently do not have glove-boxed electron-beam welding equipment for plutonium work, elements containing plutonium alloy core plates will be TIG welded. Specimen elements containing dummy core plates will be electron-beam welded. A number of dummy and fuel loaded jackets have been TIG welded. A micrographic section of one of these is shown in Fig. 21.

25X

Fig. 21. TIG Welds of End Plug and Sleeve

(vii) Inspection. Techniques of leak detection, alpha counting, and dimensional inspection previously used for ZPR-3 fuel elements appear to be satisfactory but will require modification for large-quantity production. Weld inspection, particularly, has been a problem because subjective judgements as well as measurements are required to evaluate welds. It is intended that weld-inspection standards be developed during pilot production. Methods for evaluating core-plate quality are being worked out.

b. U-Pu-based Fuel Alloys. Uranium-plutonium-based ternary alloys have been studied as alternates to the U-20 a/o Pu-6 a/o Mo that was developed for zero-power applications. Density and dilatometric measurements have been made, and all the air-corrosion tests (see Progress Report for November 1965, ANL-7122, pp. 33-34) have been continued.

The U-Pu-Mo alloy is still by far the best of the various alloys studied. With the decision to use this alloy for ZPPR fuel plates, further tests on alternative alloys have been discontinued.

Some variation has been observed in the manner in which the U-Pu-Mo alloys crack after long-time exposure to air, and it is believed that this may be related to the carbon content. Air-corrosion tests of U-Pu-Mo alloys with varying amounts of carbon added are being continued.

U-Pu-Mo alloys with plutonium contents of 25 to 30 a/o are of interest for ZPPR. Castings in this range of composition are being made and will be tested in the same manner as was the U-20 a/o Pu-6 a/o Mo alloy.

2. Metal Fuels

a. U-Pu-Zr and U-Pu-Ti Alloys

(i) Structures. Structure studies of an U-12.9 a/o Pu-22.5 a/o Zr alloy and an U-11.9 a/o Pu-25.7 a/o Ti alloy have been made. Between 1155°C (the solidus temperature) and 654°C the former alloy is a single-phase bcc gamma-uranium solid solution. During quenching to room temperature the gamma transforms to alpha uranium and a solid solution based on the delta phase in the U-Zr system. Between 654 and 596°C the alloy consists of two phases, alpha uranium and a bcc gamma phase, that is enriched in zirconium. From 596°C to room temperature, two phases are present in the alloy, alpha uranium and the previously contained delta phase.

Between approximately 1140°C (the solidus temperature) and 856°C the U-11.9 a/o Pu-25.7 a/o Ti alloy is a gamma solid solution and, as in the case of the zirconium alloy, the gamma transforms during quenching to low-temperature phases. At just below 856°C and down to 644°C the alloy consists of gamma and a ternary compound that is based on the intermediate phase U_2Ti . At lower temperatures, down to 558°C, solid solution based on the beta and eta phases in the U-Pu system appear, as based on microstructural data, to exist in equilibrium with the $(U, Pu)_2Ti$ phase. At still lower temperatures the alloy consists only of the $(U, Pu)_2Ti$ phase and zeta (U, Pu) .

The composition of the phases present in the alloys have not been determined. Preliminary findings suggest that as much as 10 a/o Pu may be soluble in the delta phase of the U-Zr system. The solubility of zirconium in the alpha, beta, and zeta phases that occur in the U-Pu systems at 560°C is restricted to less than 5 a/o.

(ii) Densities. The densities of a number of specimens of cast U-Pu-Zr and U-Pu-Ti alloys were measured. Density decreased linearly with increases of zirconium or titanium to at least 40 a/o. Plutonium in

the range of 10 to 25 a/o and small amounts of porosity did not lower the measured value more than 1%. The density was decreased as much as 10%,

TABLE VI. Density of U-Pu-Zr and U-Pu-Ti Alloys vs. the Oxygen and Carbon Impurity Content

Zr or Ti (a/o)	Density (g cm^{-3})	
	600 ppm*	0 ppm*
0	18.80	19.00
10	17.56	17.88
20	16.34	16.74
30	15.12	15.60
40	13.90	14.46

*Total carbon and oxygen impurity content.

ternary phase containing 37 a/o Zr and 31 a/o Fe. The previously observed compatibility of such U-Zr alloy with iron requires that the compositional path in diffusion layers is toward the zirconium corner of the ternary system.

however, by about 2000 ppm of oxygen and carbon impurities. Reasonable values for pure alloys have been deduced from the data and are listed in Table VI.

b. U-Zr-Fe System. The isothermal section at 800°C has been determined in order to define the compositions of phases that may occur in diffusion layers between U alloys containing more than 20 a/o Zr and pure iron. Possible phases are (i) the binary intermediate phases ZrFe_3 , ZrFe_2 , Zr_2Fe , and Zr_3Fe ; (ii) bcc solid solution of U and Zr containing less than 5 a/o Fe; (iii) a

3. Jacket Development

Creep tests are being conducted on 0.5-mm-thick V-20 w/o Ti sheet specimens that have been pre-exposed to sodium at 650°C in a dynamic loop. (Vacuum distillation analyses on two sodium samples yielded oxygen contents of 18 and 20 ppm.) Some results are shown in Fig. 22. Curve A represents the creep of a specimen pre-exposed to sodium for seven days. Curve B represents a control specimen-vacuum encapsulated in Vycor and heated for seven days at 650°C . Curve C represents a thicker, 1.5-mm sheet specimen not exposed to sodium. All specimens were in the 900°C recrystallized condition. Short-time exposure to liquid sodium apparently decreases creep resistance.

Fig. 22
Effect of Sodium on V-20 w/o Ti.
See text for description of curves.

4. Corrosion

a. Fuel-cladding Materials for Sodium-cooled Reactors. Samples of V-20 w/o Ti have been exposed to sodium containing 3 to 5 ppm oxygen (measured by vacuum distillation) at temperatures of 700 and 750°C for periods of time up to 32 hr. Samples are rotated at about 5 rpm to insure access of oxygen to the surface. The oxidation at 750°C is parabolic with a parabolic rate constant $K_p = 43 \times 10^{-4} \text{ mg}^2/\text{cm}^4\text{-hr}$ for the 32-hr period of exposure.

At 700°C the process is linear for the first 12 hr and is followed by a transition to a parabolic loss for the balance of the exposure of 32 hr. The respective rate constants are: $K_l = 8 \times 10^{-3} \text{ mg}/\text{cm}^2\text{-hr}$ and $K_p = 13 \times 10^{-4} \text{ mg}^2/\text{cm}^4\text{-hr}$.

Exposure of V-20 w/o Ti at 600°C is in progress.

b. Refractory Metal Alloys for Service in Oxygen-contaminated Sodium. Further efforts have been made to improve the poor fabricability of V-20 w/o Cr-10 w/o Al and retain its much improved (over V-20 w/o Ti) corrosion resistance. Small levitation melts of the V-20 w/o Cr-10 w/o Al basic composition with additions of 1-2 w/o Zr or 0.5-1 w/o Y have been prepared for fabricability evaluation.

A range of alloys was also prepared by levitation melting in which part or all of the chromium in V-Cr-Al was replaced by titanium. These alloys were exposed for one week to 650°C sodium containing about 25 ppm oxygen. The comparison of external loss is shown in Table VII.

TABLE VII. Corrosion of Vanadium Alloys in 650°C Sodium for 7.0 days

Alloy* (w/o)	External Loss (μ per side)
V-20 Ti (Reference)	33
V-30 Ti-10 Al	25
V-20 Ti-7 $\frac{1}{2}$ Al	31
V-20 Ti-10 Al	27
V-10 Ti-10 Al	45
V-10 Ti-10 Al-10 Cr	19

*Alloys (except V-20 w/o Ti reference) exposed in the as-cast, machined, and etched condition. V-20 Ti in the 900°C-1 hr annealed and etched condition.

It is obvious that the corrosion resistance of these alloys does not approach that of V-20 w/o Cr-10 w/o Al, which experienced a 3 μ external loss under the same conditions (see Progress Report for November 1965, ANL-7122, p. 35).

Samples of V-15 w/o Ti-5 w/o Cr were also exposed to sodium for one week with the results shown in Table VIII. In this case the heat treatments at higher temperature led to a significant reduction of the dimensional loss of the samples.

TABLE VIII. Corrosion of V-15 w/o
Ti-5 w/o Cr in Sodium at 650°C
Containing 25 ppm Oxygen

Condition	External Loss (μ per side)
As rolled	40
Annealed at 700°C for 1 hr	31
Annealed at 900°C for 1 hr	31
Annealed at 1350°C for 1 hr	24
Annealed at 1500°C for 1 hr	13

The dynamic test facility has been completed and successfully tested. An initial loading of V-20 w/o Ti suffered erosive attack during a one-week exposure to sodium at 650°C with a nominal oxygen content of 20 ppm and flowing at a speed of 6 m/sec. Analysis of the sodium for oxygen is under way, and a duplicate experiment has been scheduled.

In presenting the corrosion data for various vanadium-base alloys in the November 1965 Monthly Report (ANL-7122), the times of exposure were inadvertently omitted from Tables VIII and IX (pages 34 and 35). In both cases the results apply to one-week tests.

5. Irradiation Testing

a. Irradiated EBR-II Blanket Rods. The uranium slugs from four upper blanket rods from subassembly C-140, four from C-143, and three lower blanket rods from C-104 were examined for evidences of dimensional changes. The fuel in these subassemblies had been irradiated to maximum calculated burnups of 0.26, 0.48 and 1.23 a/o, respectively, corresponding to ~0.04 a/o maximum burnup in the depleted uranium blanket metal. Although little dimensional change would be expected from this low burnup level, there is the possibility that thermal cycling could cause elongation of the rolled and beta-quenched blanket slugs. No significant changes in dimensions (length and diameter) and density were observed. The density values from end to end of the slugs were scattered at 0.26 and 0.48 a/o fuel burnup, but were quite uniform for each slug in the 1.23 a/o fuel burnup subassembly.

b. Irradiated EBR-II Fuel Cladding. As part of the surveillance program of irradiated fuel rods from EBR-II, tube-burst tests are being made for Type 304 stainless steel jackets. The jackets are long tubes closed at one end with a welded plug. A spiral spacer wire (of 1.2-mm dia) is spot welded to the bottom plug and to the tube near the top. The bottom plug external to the tube is in the shape of a two-pronged fork. The tube itself, as initially tested, is about 44.5 cm long and of 4-mm ID with a 230- μ wall. The initial overall length is about 46 cm.

The jackets are removed from the fuel in the alpha-gamma cave in a nitrogen atmosphere to reduce the extent of oxidation of sodium. The temperature of the fuel rod was raised to 200°C to melt the sodium. After the melting it was possible to remove the fuel and the bulk of the sodium with relative ease. No attempt was made to remove the sodium from the jackets except by melting.

The test specimens are connected to pressure tubing on the cover of the burst-test chamber while in the nitrogen cell, evacuated, and filled with pressurized argon or helium for bursting. The temperature along the entire length of the specimen is held to $\pm 5^\circ\text{C}$ of the set temperature. In some of the later tests the tube at the connection was kept several degrees cooler in an attempt to promote rupture of the irradiated specimens away from the vicinity of the connection. The fuel jackets tested to date are described in Table IX.

TABLE IX. Identification of Irradiated Fuel-rod Jackets

Subassembly	Fuel Rod	Radial Distance from Core Center (cm)	Exposure (MWd)	Maximum Fast Neutron Flux at 62.5 MW* ($\text{n}/\text{cm}^2\text{-sec} \times 10^{-15}$)	Maximum Total Fast Flux ($\text{n}/\text{cm}^2 \times 10^{-22}$)
S-602	E26	14.26	2927	2.6	1.1
S-602	E36	9.38	2927	3.1	1.3
C-104	E2	12.93	3320	2.8	1.3
C-104	E36	10.54	3320	3.1	1.4
C-104	E90	7.60	3320	3.3	1.5

*ANL-5719 (Addendum). Figures 38 and 39 on pp. 142-143 for core midplane.

The maximum stress (tangential or circumferential stress at the inner surface of the tube) at rupture is calculated by using the expression

$$\sigma_{\max} = P \frac{K^2 + 1}{K^2 - 1},$$

where K is the ratio of the outside diameter to the inside diameter of the tube. Typical data are given in Table IX. The calculated stresses have been plotted against the test temperatures in Fig. 23 for both the irradiated and unirradiated specimens.

Fig. 23. Rupture Stress vs. Test Temperature

A large difference exists between the maximum stress for the irradiated versus the unirradiated specimens, but this difference decreases with increasing temperature.

Diameter measurements along the length of the tube have been used to provide a measure of elongation. Since no measurements were possible at the locale of the rupture, the elongation percentages given in Table X correspond more nearly to "uniform elongation." The tubes at the rupture were badly torn and free fragments often resulted. None of the irradiated

specimens that broke near the pressure connection showed ductility greater than 1% elongation. Jacket S-602-E26, which broke at the end away from the connection, had a diameter increase equivalent to 5% elongation next to the rupture, but less than 1% elsewhere along the length. The unirradiated specimens showed average elongations of 9.6 to 12.7%. A summary of observations to date is as follows:

- (i) The tube rupture strength of irradiated specimens is much greater than that of unirradiated specimens in the test temperature range from 400 to 550°C.
- (ii) The rupture strength of both the irradiated and unirradiated tube specimens decreases with increasing temperature in the range from 400 to 550°C.
- (iii) The rupture strength of the irradiated specimens decreases at a faster rate with increasing test temperature than do the unirradiated.
- (iv) Hardness measured at room temperature increases in going from the top of the jacket toward the middle of the length.
- (v) The rupture strength at 500°C increases in going from the top of the jacket towards the middle of the length.
- (vi) Ductility of the irradiated specimens is low.
- (vii) Welding of the spacer wire to the tube has not resulted in an area of weakness.

TABLE X. Burst-test Data

Specimen	Test Temperature (°C)	Burst Pressure (psi)	Tangential Rupture Stress (psi)	Elongation (%)		
				Max	Min	Avg
<u>Irradiated</u>						
S-602-E36	400	8875	81,600	1*	-	-
C-104-E90	450	8252	75,900	1*	-	-
C-104-E2	500	7525	69,200	1*	-	-
C-104-E2	500	8425	77,500	0.7*	-	-
C-104-E2	500	8675	79,700	0.6	-	-
C-104-E2	500	2825	-	-	-	-
S-602-E26	500	6850	63,000	5.1*	-	-
C-104-E36	550	6875	63,200	0.5*	-	-
<u>Unirradiated</u>						
-	300	5800	53,300	12.3	9.9	11.2
-	400	5425	49,900	12.7	7.7	10.9
-	450	5400	49,600	14.3	10.8	12.7
-	500	5200	47,800	11.6	8.2	9.6
-	500	5275	48,500	13.0	9.1	10.5
-	550	4925	45,300	14.4	8.8	11.2

*Near break. Values of other measurements of diameter are lower so that no adequate indication of elongation is possible.

c. Irradiation Program for Fast Reactors. The Laboratory's program of irradiation of fast reactor materials includes metallic and ceramic fuels in various fuel-jacket alloys, and test specimens of the jacket alloys themselves. The reactors being used for the irradiations include CP-5, MTR, and EBR-II.

(i) Metallic Fuel Irradiations. Irradiations of 63 specimens under way are shown in Table XI. The specimens being irradiated in CP-5

TABLE XI. Status of Metal Fuel Irradiations in Progress

Specimen No.	Reactor	Fuel Composition (w/o)	Clad Composition (w/o)	Clad ID (mm)	Clad thickness (mm)	Maximum Clad Temp (°C)	Calculated Burnup to Date	
							a/o	/cm ³ × 10 ⁻²⁰
N-15	CP-5	U-19 Pu-14 Zr	V-20 Ti	4.4	0.38	560	5.0	16.5
ND-23	EBR-II	U-15 Pu-10 Zr	V-20 Ti	4.5	0.41	540	1.08	3.97
ND-24	EBR-II	U-15 Pu-10 Zr	V-20 Ti	4.5	0.41	535	1.08	3.97
ND-28	EBR-II	U-15 Pu-9 Zr	304 SS	4.2	0.51	650	0.53	2.00
ND-41	EBR-II	U-15 Pu-9 Zr	304 SS	4.2	0.51	650	0.53	2.00
ND-32	EBR-II	U-15 Pu-9 Zr	316 SS	4.2	0.38	645	0.53	2.00
ND-43	EBR-II	U-15 Pu-9 Zr	Hastelloy-X	4.2	0.38	645	0.53	2.00
ND-25	EBR-II	U-14 Pu-12 Zr	304 SS	4.2	0.51	630	0.47	1.64
ND-27	EBR-II	U-14 Pu-12 Zr	304 SS	4.2	0.51	630	0.50	1.74
ND-26	EBR-II	U-14 Pu-12 Zr	316 SS	4.2	0.38	625	0.47	1.64
ND-29	EBR-II	U-14 Pu-12 Zr	316 SS	4.2	0.38	625	0.47	1.64
ND-30	EBR-II	U-14 Pu-12 Zr	316 SS	4.2	0.38	635	0.50	1.74
ND-31	EBR-II	U-14 Pu-12 Zr	316 SS	4.2	0.38	635	0.50	1.74
ND-33	EBR-II	U-14 Pu-12 Zr	Hastelloy-X	4.2	0.38	630	0.50	1.74
ND-34	EBR-II	U-14 Pu-12 Zr	Hastelloy-X	4.2	0.38	635	0.50	1.74
ND-35	EBR-II	U-14 Pu-12 Zr	Hastelloy-X	4.2	0.38	640	0.50	1.74
ND-37	EBR-II	U-14 Pu-12 Zr	Hastelloy-X-280	4.5	0.38	635	0.50	1.74
ND-39	EBR-II	U-14 Pu-12 Zr	Hastelloy-X-280	4.5	0.38	635	0.50	1.74
ND-44	EBR-II	U-14 Pu-12 Zr	Hastelloy-X-280	4.5	0.38	635	0.50	1.74
N-14	CP-5	U-15 Pu-10 Ti	V-20 Ti	4.4	0.38	560	5.0	16.8
NC-17	EBR-II	U-15 Pu-10 Ti	V-20 Ti	4.5	0.41	540	1.08	3.64
NC-23	EBR-II	U-15 Pu-10 Ti	V-20 Ti	4.5	0.41	540	1.09	3.68
N-10	CP-5	U-10 Pu-10 Fz	V-20 Ti	4.2	0.41	530	4.8	18.2
N-11	CP-5	U-10 Pu-10 Fz	V-20 Ti	4.1	0.41	530	4.8	18.2
N-12	CP-5	U-10 Pu-10 Fz	V-20 Ti	4.0	0.41	530	4.8	18.2
CK-01	MTR	U-10 Pu-10 Fz	Nb-1 Zr	4.0	0.38	555	3.3	1.25
CG-02	EBR-II	U-10 Pu-10 Fz	Nb-1 Zr	4.0	0.23	445	0.42	1.60
CG-03	EBR-II	U-10 Pu-10 Fz	Nb-1 Zr	4.0	0.23	440	0.41	1.56
CJ-01	EBR-II	U-10 Pu-10 Fz	Nb-1 Zr	4.0	0.23	435	0.40	1.52
PB-02	EBR-II	U-10 Pu-10 Fz	Nb-1 Zr	4.0	0.23	440	0.41	1.56
TC-1	CP-5	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	500	1.9	4.7
C-152P	CP-5	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	500	1.9	4.7
C-153P	CP-5	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	500	1.9	4.7
C-155P	CP-5	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	540	1.9	4.7
C-156P	CP-5	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	540	1.9	4.7
C-159P	CP-5	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	540	1.9	4.7
N-7	CP-5	U-15 Pu-10 Fz	V-20 Ti	4.2	0.41	530	4.8	18.2
N-8	CP-5	U-15 Pu-10 Fz	V-20 Ti	4.1	0.41	530	4.8	18.2
N-9	CP-5	U-15 Pu-10 Fz	V-20 Ti	4.0	0.41	530	4.8	18.2
N-13	CP-5	U-15 Pu-10 Fz	V-20 Ti	4.3	0.38	500	3.9	14.8
CE-03	MTR	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.38	555	3.3	1.3
EC-01	MTR	U-15 Pu-10 Fz	V	4.0	0.38	555	3.3	1.3
CA-01	EBR-II	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	450	0.41	1.56
CB-02	EBR-II	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.38	445	0.42	1.60
CB-03	EBR-II	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.38	440	0.41	1.56
CB-04	EBR-II	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.38	440	0.41	1.56
CD-01	EBR-II	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	440	0.41	1.56
CD-02	EBR-II	U-15 Pu-10 Fz	Nb-1 Zr	4.0	0.23	430	0.40	1.52
LA-02	EBR-II	U-15 Pu-10 Fz	Hastelloy-X	3.9	0.25	430	0.40	1.52
PA-01	EBR-II	U-15 Pu-10 Fz	Nb-4 V	4.0	0.23	435	0.40	1.52
F-30	CP-5	U-15 Pu-10 Fz	304 SS	3.9	0.38	570	3.2	12.1
F-31	CP-5	U-15 Pu-10 Fz	304 SS	3.9	0.38	570	3.2	12.1
F-32	CP-5	U-15 Pu-10 Fz	Hastelloy-X	4.2	0.23	570	3.2	12.1
F-33	CP-5	U-15 Pu-10 Fz	304 SS	4.2	0.38	610	3.2	12.1
L-16	CP-5	U-15 Pu-10 Fz	304 SS	4.2	0.38	610	3.2	12.1
L-17	CP-5	U-15 Pu-10 Fz	Hastelloy-X	4.2	0.23	610	3.2	12.1
C-93	EBR-II	U-20 Pu-10 Fz	Nb-1 Zr	4.0	0.23	435	0.41	1.56
C-97	EBR-II	U-20 Pu-10 Fz	Nb-1 Zr	4.0	0.23	430	0.40	1.52
C-98	EBR-II	U-20 Pu-10 Fz	Nb-1 Zr	4.0	0.23	430	0.39	1.48
C-99	EBR-II	U-20 Pu-10 Fz	Nb-1 Zr	4.0	0.23	430	0.40	1.52
C-100	EBR-II	U-20 Pu-10 Fz	Nb-1 Zr	4.0	0.23	430	0.40	1.52
C-101	EBR-II	U-20 Pu-10 Fz	Nb-1 Zr	4.0	0.23	435	0.40	1.52
CM-01	EBR-II	U-20 Pu-10 Fz	Nb-1 Zr	4.0	0.38	435	0.40	1.52

are from 10 to 15 cm long and are in instrumented capsules. Those in MTR are full-length EBR-II size (46 cm long) and are in instrumented, temperature-controlled capsules. The specimens in EBR-II are also full-length and are located in three special subassemblies. Most of the fuel alloys under irradiation are of the uranium-plutonium-fissium type because of their relevance to the EBR-II pyrometallurgical reprocessing cycle. More recently, greater interest has developed in high-melting U-Pu-Zr and U-Pu-Ti alloys of uranium and plutonium. Specimens of these alloys will replace most of the fissium-type alloys in the irradiation program.

The CP-5 reactor resumed operation on December 16, 1965 after a four-month shutdown for modification of a thermal column. Six irradiation capsules containing metallic fuel specimens were installed in CP-5 vertical thimbles prior to this startup. The capsules containing specimen No. N-14 (U-15 w/o Pu-10 w/o Ti) was removed from the reactor because of thermocouple failures. The capsule will be neutron radiographed to determine the condition of the specimen. If its condition is satisfactory, irradiation will be resumed. The capsule containing specimens F-30, F-31, F-32, F-33, L-16, and L-17 was removed from the reactor because of thermocouple indications of possible jacket failure. The capsule will also be neutron radiographed for examination of the experimental specimens.

(ii) Ceramic Fuel Irradiations. The irradiations in progress in EBR-II are summarized in Table XII. Two types of UC-20 w/o PuC are being studied: physically mixed powders of UC and PuC in the form of vibratorily compacted rods, and solid solution (U-Pu)C in the form of both vibratorily-compacted powder and sintered pellets. The effective or "smeared" density of all rods ranges from 80 to 85% of theoretical. The irradiations in EBR-II on full-length rods (36-cm-long fuel sections) are being closely coordinated with those of smaller specimens in instrumented capsules for the High-temperature Fuels Program.

TABLE XII. Status of Ceramic Fuel Irradiations in EBR-II

Specimen No.	Fuel* Material	Fuel Form	Jacket Material	Max Jacket Temp (°C)	Burnup	
					a/o	fiss/cc x 10 ²⁰
SMV-2	UC-20 w/o PuC	Vipac	304 SS	645	1.4	3.7
HMV-5	UC-20 w/o PuC	Vipac	Hastelloy-X	670	1.4	3.7
NMV-11	(U _{0.8} Pu _{0.2})C	Vipac	Nb-1Zr	645	1.4	3.7
NMP-2	(U _{0.8} Pu _{0.2})C	Pellet	Nb-1Zr	545	0.33	0.86
NMV-4	UC-20 w/o PuC	Vipac	Nb-1Zr	635	0.47	1.2
NMV-7	UC-20 w/o PuC	Vipac	Nb-1Zr	605	0.43	1.1
NMV-12	(U _{0.8} Pu _{0.2})C	Vipac	Nb-1Zr	635	0.47	1.2
HMV-1	UC-20 w/o PuC	Vipac	Hastelloy-X	640	0.42	1.1
HMV-4	UC-20 w/o PuC	Vipac	Hastelloy-X	670	0.46	1.2
HWMP-1	(U _{0.8} Pu _{0.2})C	Pellet	Hastelloy-X ⁽¹⁾	555	0.32	0.84
HWMV-1	(U _{0.8} Pu _{0.2})C	Vipac	Hastelloy-X ⁽¹⁾	685	0.47	1.2

*Mixed carbides are identified as UC-X w/o PuC and solid solution carbides as (U_xPu_z)C.

⁽¹⁾Has 0.13-mm layer of vapor-deposited tungsten on the ID.

A subassembly is now being prepared in which rods of mixed carbides, similar to those above, and mixed oxides will be irradiated in EBR-II. The UO_2 -20 w/o PuO_2 rods are vibratorily compacted Nupac powder supplied by Battelle Northwest Laboratories. Jacket materials are Types 304 and 316 stainless steel and vanadium.

6. Supplemental U-5 w/o Fs Fuel Pins for EBR-II

Approximately 200 kg of U-5 w/o Fs fuel pins were needed to supplement the Fuel Cycle Facility production for EBR-II. This quantity of material will produce about 2800 pins of 3.66-mm dia and 343 mm long. The alloy was prepared and cast into 6.5-kg billets. The fuel pins are being injection cast into precision Vycor molds in the Plutonium Fabrication Facility where the necessary equipment was available in operation. Since the glovebox lines are contaminated with plutonium, the fuel pins are handled as " α -radioactive" on all subsequent operations.

A total of 2356 fuel pins have passed inspection by January 28, 1966. Of these 2131 have been shipped to Idaho for reactor use; 343 were 70 w/o U^{235} enriched, and 1513 were 52 w/o U^{235} enriched.

7. Development of Nupac-type Fuel Elements

a. Particulate Fuel Test Specimens. The final design was set for 23 fuel rods containing UO_2 -20 w/o PuO_2 processed by the Battelle-Northwest Nupac technique. The rods are to be fabricated by vibratory compaction, jacketed in Type 304 stainless steel, Hastelloy-X, and vanadium-20 w/o titanium, and to contain fuel densities of 80-82, 83-85, >85% theoretical.

Twelve rods have been fabricated. Characteristics of the completed fuel specimens are presented in the following:

Rod No.	Rod Composition in Grams					Percent Theoretical Density	Jacket Material
	U^{235}	U^{238}	Pu^{239}	Pu^{241}	Total Oxide		
SOU-1	55.74	3.08	13.69	0.12	85.05	79.9	304-SS
SOU-2	55.74	3.08	13.69	0.12	85.05	79.8	304-SS
SOU-3	58.03	3.21	14.26	0.12	88.55	83.1	304-SS
SOU-4	57.25	3.16	14.06	0.12	87.35	82.1	304-SS
SOU-5	58.10	3.21	14.27	0.12	88.65	83.4	304-SS
SOU-6	58.08	3.21	14.27	0.12	88.61	83.3	304-SS
HOU-1	60.40	3.54	14.84	0.13	92.20	80.0	Hastelloy-X
HOU-2	60.38	3.54	14.84	0.13	92.17	79.9	Hastelloy-X
HOU-3	60.40	3.54	14.84	0.13	92.20	79.9	Hastelloy-X
HOU-4	60.41	3.54	14.85	0.13	92.21	80.0	Hastelloy-X
HOU-15	60.11	3.52	14.77	0.13	91.75	79.8	Hastelloy-X
HOU-6	60.39	3.54	14.84	0.13	92.18	80.1	Hastelloy-X

D. General Fast Reactor Fuel Reprocessing Development

1. Skull Reclamation Process

The skull-reclamation process is being developed for the recovery and purification of uranium in a residue or skull that is left in the crucible after melt refining of fuel pins discharged from the EBR-II reactor. The amount of uranium in the crucible is 5 to 10% of the original uranium charge. The chemistry and technology developed for the skull reclamation process has considerable potential for a wide variety of uranium-bearing fuels.

Four reduction-furnace runs (SRR-11 through -14) have been completed (see Progress Report for December 1965, ANL-7132, pp. 23-24) under more vigorous reaction conditions (higher temperatures, longer contact times, and faster mixing) than in earlier runs. The noble-metal-extraction step was carried out for 2 hr at 750°C. The reduction step was conducted for 2 hr, of which 3/4 hr was required to heat the charge from 700°C to the operating temperature of 800°C. Mixing speed in both steps was 800 rpm. This new series of runs is designed to verify the adverse effect of fluoride ion on fission product decontamination in the noble metal extraction step (see Progress Report for November 1965, ANL-7122, p. 39) and the beneficial effect of fluoride ion on the reduction of uranium oxides in the reduction step. Accordingly, fluoride ion, when used, was added to the salt between the noble metal extraction and reduction steps. (The process steps were previously shown in the flowsheet in Progress Report for June 1965, ANL-7071, p. 24.)

In runs SRR-11 and SRR-12, high removals of molybdenum and ruthenium were again demonstrated, ranging from ~60% to ~90%. These results are very satisfactory and confirm those previously obtained (see ANL-7122, p. 38). The fission product removals for runs SRR-13 and -14 are not yet available. Overall fission product removals will be determined by analyses of the uranium buttons obtained after retorting the zinc-magnesium-uranium product solutions.

Excellent reductions of uranium oxides (~99%) have been realized in two runs (SRR-13 and -14) in which 10 m/o fluoride ion was added as CaF_2 just prior to the reduction step. In SRR-11, no fluoride ion was used in the extraction and reduction steps, and the reduction of uranium oxide was 93% complete; in SRR-12, 2 m/o fluoride ion was added as NaF just before the reduction step, and the oxide reduction was 96% complete. The beneficial effect of fluoride ion in the reduction step appears to have been confirmed in these runs.

Mechanical operations, namely, charging of materials, heating, mixing, sampling, and transferring molten metals and fluxes from the furnace to exterior receivers, were performed without difficulty. The fume-control

system, which is connected to the furnace-charging port and the transfer receiver, satisfactorily prevented the escape of metal and salt fumes (see ANL-7132, p. 24) to the inert atmosphere enclosure in which the reduction furnace is installed.

Two plant-scale retorting runs (PSR-8 and PSR-9) have been completed with Zn-Mg-U ingots from reduction-furnace runs SRR-11 and SRR-12. The runs were conducted in a thixotropically cast beryllia crucible that had been used in five previous retorting runs. In the retorting step, the magnesium-zinc is distilled under vacuum (~ 10 Torr) at 650 to 750°C from the uranium product solution, after which the uranium is melted ($\sim 1200^\circ\text{C}$) to form an ingot.

In run PSR-8, the Zn-Mg-U ingot from run SRR-11 was retorted without removing the small amount of flux which has been transferred along with the ingot. This resulted in a uranium product that contained a large quantity of oxide. Prior to carrying out run PSR-9, flux adhering to the ingot from run SRR-12 was removed by dissolution with water, after which the ingot was rinsed with acetone and then allowed to dry. Retorting of this ingot resulted in a clean uranium product. Further study is planned for developing a method (the above procedure is not appropriate for actual operation) for separating flux from the Zn-Mg-U solution during its transfer from the reduction furnace.

2. Processes for Future Fast Reactor Fuels

Compact pyrochemical processes are under development for processing fast breeder reactor fuels of the ceramic (e.g., oxide or carbide) or metallic types. Emphasis is currently being placed on the development of salt-transport processes, which have the inherent advantage over other pyrochemical processes that only small solution volumes are required, since not all of the uranium or plutonium needs to be in solution at any one time. Such processes also offer the possibility of excellent uranium-plutonium-fission product separations and excellent recoveries of uranium and plutonium (see Progress Report for December 1965, ANL-7132, pp. 24-28).

To date, most of the experimental work has been carried out with liquid copper-magnesium and zinc-magnesium alloys in mutual contact with molten MgCl_2 at 800°C . With these alloys, uranium is transferred from the Cu-Mg phase through the salt phase into the Zn-Mg phase. Simultaneously, magnesium is transferred to the Cu-Mg phase. The copper-magnesium alloys offer the advantages of a reasonably good single-stage separation of rare earths from plutonium, good uranium and plutonium transfer rates, and internal control of the magnesium concentration in the Cu-Mg alloys through the dissolution of excess solid copper in the system as magnesium is transferred into a Cu-Mg alloy.

Laboratory-scale experiments have established the uranium solubility in Cu-Mg alloy. Uranium solubility decreases smoothly from 3.42 w/o in Cu-9 w/o Mg alloy to 0.034 w/o in Cu-77 w/o Mg alloy. At magnesium concentrations of 6 to 10 w/o in the Cu-Mg alloy, the uranium distribution coefficient and uranium solubility are such that a uranium concentration of 1 to 2 w/o can be obtained in the molten MgCl_2 in contact with the uranium-saturated Cu-Mg alloy. This solubility would result in a fairly rapid uranium transfer.

Another alloy being considered for the salt transport process is cadmium-copper-magnesium, which offers the possibility of lowering the operating temperature to $\sim 600^\circ\text{C}$ while still retaining good separation of uranium and plutonium from rare earths. In an exploratory study, the solubility of uranium in Cu-Cd alloys (zero magnesium) was determined. At a temperature of 600°C , the uranium solubility is about 1.9 w/o in Cu-85 w/o Cd alloy and about 0.5 w/o in Cu-67 w/o Cd alloy. These solubilities appear to be adequate for process use. Measurements will be extended to systems containing magnesium. An estimate of the phase diagram for this ternary system (Cu-Cd-Mg) in the region of process interest indicates that control of the magnesium concentration can probably be accomplished by the same technique as used with the copper-magnesium system, i.e., by dissolution of excess copper as magnesium transfers in.

A Cd-Zn-Mg alloy, which would also permit low-temperature (500 - 600°C) operation, is also being considered. A zinc-cadmium alloy, to which a small amount of magnesium was added to assist in dissolution of the uranium (via reduction of oxide films), was first saturated with uranium and then equilibrated with a 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl salt. The resultant uranium concentrations in the salt ranged from 9.5 w/o at 650°C to 1.8 w/o at 448°C . At low magnesium concentrations, the uranium solubility and distribution coefficient relationships in this metal/salt system are such as to provide rapid uranium transfer. However, the transfer rate is extremely sensitive to magnesium concentration, a marked change occurring with only a slight variation in magnesium concentration. Consequently, methods for controlling the magnesium concentration at the necessary low level are actively being sought. With this system, a multistage separation would be required for an adequate removal of rare earth fission products from plutonium.

3. Materials Evaluation

A supporting program to find inexpensive, easily fabricated containment materials for pyrochemical process solutions is underway. Type 405 stainless steel is one of the more promising alloys for use with cadmium-based systems containing low percentages of magnesium (see Progress Reports ANL-7090, August 1965, pp. 28-29, ANL-7045, April 1965, p. 23, and ANL-7003, January 1965, pp. 20-21). The stability of a 0.45 w/o solution of uranium in cadmium-3.7 w/o magnesium alloy on long-term (169-hr)

exposure at 425°C to Type 405 stainless steel was recently determined. (A 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl molten-salt phase was also present in the experiment.) After an initial loss of 3.2% of the uranium from the liquid metal solution, no further uranium loss occurred during the remainder of the 169-hr test period. Thus it appears that Type 405 stainless steel is a satisfactory container material for this system, after it has undergone an initial conditioning exposure to the solutions.

Croloy 16-1, a readily fabricated ferritic stainless steel similar to Type 405 stainless steel, corroded at a fairly high rate in a Cu-10 w/o Mg-2 w/o U alloy and in a Mg-35 w/o Zn-1 w/o U alloy at 800°C (~2 mil/24 hr for both alloys in the presence of a 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl salt). Consequently, this material could only be used in applications where contact with the metal/salt solvent system is for a limited time, such as transfer lines. On the other hand, cast iron crucibles exhibited no detectable corrosion or deterioration on exposure to the Cu-10 w/o Mg-2 w/o U/salt system for 170 hr at 800°C or to a Cd-8 w/o Zn-4 w/o U/salt system for 290 hr at 600°C. More extensive tests of cast iron are therefore planned.

Tungsten is the most satisfactory material for containment of high-zinc alloys. A previous test of an agitator blade, fabricated of pressed and sintered tungsten containing 0.1 w/o nickel to reduce porosity of the finished tungsten shape,⁴ gave no evidence of deterioration on exposure to Zn/47.5 m/o MgCl_2 -47.5 m/o CaCl_2 -5 m/o MgF_2 at 800°C for 200 hr (Progress Reports August 1965, ANL-7090, p. 29, and November 1965, ANL-7122, p. 41). Testing of this material was extended by exposure of additional coupons, as agitator blades, which were used to stir a Zn/50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl system for 380 hr at 800°C. No appreciable corrosion or structural changes were observed. It is concluded that a small nickel addition to the tungsten does not significantly affect the corrosion properties in zinc/salt systems and that tungsten with 0.1 w/o nickel would be acceptable for process requirements.

E. Sodium Technology

1. Components Surveillance Loop

a. Construction. Additional work has been performed in the construction of the loop (see Progress Report for July 1965, ANL-7082, pp. 37-38). The electromagnetic (EM) pump has been assembled and welded into the system. Piping and the Conoseal connection flanges have been installed; site locations for instrumentation has been selected. Arrangements have been made to connect the EM pump to two high-current rectifiers.

b. Modified Falex Wear Tester. The first model of the modified Falex wear tester to be used in the components surveillance loop has been tested with very satisfying results.

⁴Oustalet, J. A., Oak Ridge National Laboratory, private communication.

In the first test a Type 304 stainless steel shaft was rotated without lubrication between bearing blocks of the same material; as expected, severe galling developed. The effect was reflected in the current response to the d.c. drive motor, which demonstrated that the onset of galling can be recorded by a measurement of the motor current.

In a second test, a carbon steel shaft was mounted between blocks of Type 304 stainless steel. The unit was operated dry and

lubricated with either water or #10 and #30 motor oil. The results are shown in Fig. 24. Both expansion and contraction were recorded through measurement of positional movement by the linear differential transformer. It can be seen from Fig. 24 that progressive wear results in a shift of the line toward the right side of the chart.

Fig. 24. Falex Wear Test

With the shaft rotating at 80 rpm and with a loading equal to 5500 psi, the friction caused a generation of heat, resulting in the block and shaft expansion. In addition, the oil lubricant film was also pumped into the contact region by the shaft and the resultant expansion is recorded by the device. When the motor was stopped, the shaft cooled and the oil film decreased, as shown

by the displacement toward the right. When the motor was restarted, heating and pumping reoccurred. The influence of start-stop wear is clearly shown in the figure.

The new equilibrium position taken upon restart shows a progressive incremental shift toward the direction of increased wear, which is greater than for the steady running condition. This is attributed to the "dry start" situation which results when the oil film has been squeezed down and new oil was not pumped in to replenish the film.

When water was used to replace the oil, the superior cooling characteristics of the water produced an immediate contraction in the components. Consequently, as the system was operated, the expansion or film pumping was not as great as with the oil. Therefore, the chart does not show any large deflections. However, the wear rate was appreciable since the lubrication properties of the water are poor. When #30 oil was substituted, the heating expansion reappeared, but the film thickness was also a little greater than the one produced by the #10 oil.

These tests have demonstrated the capability of the device to reflect quantitatively the phenomena of rubbing contact. To study the behavior of mechanisms in sodium, this unit will be a very useful tool when applied to the 1200°F materials and component testing loop.

2. Dynamic Components Test Loop

The grid plate has been leveled and installed in the test vessel. The Conex flange on the sodium inlet line to the vessel was welded in place, and X-raying indicated a sound weld.

Preparations for strain test of the system for piping sodium have continued (see Progress Report for November 1965, ANL-7122, p. 11). Since the piping insulation covering has been completed, the external wiring connections of the strain gauges and thermocouples was started. The next task is to connect each gauge to its proper terminals of the reading instruments and provide necessary insulation to the leads.

Calrod heaters located on the pipe to the test vessel inlet and strain gauge mounting on the loop pipe section were replaced after welding the Conex fitting to the ID of the sodium inlet.

The entire system is now under vacuum for a field leak check of all recently completed welds.

The test-vessel spool piece, for access to instrumentation leads, is gasketed and bolted in place.

Lagging of the large sodium pump is now completed.

A 7.5-kW emergency generator has been provided.

After completion of three more internal welds on the sodium inlet pipe, the cold-trap circuit, requiring two additional welds, will be tied into the large main loop. This will complete the mechanical construction.

3. Small Static Soak Pots

Small pots are being used as a soaking facility for self-welding tests. The field assembly of the first test vessel is completed and has been filled with sodium. The test vessel is now at 1200°F with no load on the samples. A strain-gauge "load cell" is being calibrated so that the force necessary to separate the metal self-welding samples may be measured accurately.

4. Evaluation of Type 304 Stainless Steel Tubing

Screening studies have been conducted to show the effect of sodium, air, and inert gas on Type 304 stainless steel tubing used for EBR-II blanket elements. Preliminary results are as follows:

(a) By measurement of rupture pressures for various lengths, a minimum length-to-diameter (L/D) ratio to yield a constant burst pressure has been derived. For a 0.493-in.-OD x 0.014-in.-wall tube, the minimum length of tubing was determined to be 3.0 in., that is, an L/D ratio of 6.

(b) Cold (room temperature) burst pressures of 20 samples of tubing averaged about 5950 psig, corresponding to a stress to rupture of 106,000 psi. Yield points and ultimate tensile strength were 34,600 psi and 110,000 psi, respectively.

(c) Cold yield point, rupture pressure, and ultimate strength were measured on tubular samples heated in air at 1200°F for seven days and heated in sodium at 1200°F for seven days. Yield points of air-heated samples were 37,000 psi as compared with 41,000 psi for sodium-soaked samples.

Ultimate strength for air-heated samples was 120,000 psi, the same as obtained for sodium-soaked samples.

Cold burst pressures were 114,000 psi for the air-heated samples and 111,000 psi for the sodium-soaked samples.

(d) Burst pressure (cold) for one sample tube subjected to 10,000 cycles at room temperature and 70% of yield strength was 104,000 psi. Cold burst pressure for a sodium-soaked sample also cycled 10,000 times at room temperature and at 70% yield strength was 105,000 psi.

The hot burst rupture strength of one sample was 34,000 psi; a sodium-soaked sample measured 37,000 psi.

(e) A Strauss test was used to determine the degree of sensitization of air-, argon-, and sodium-treated samples to compare with as-received EBR-II blanket tubing. In a 2.0-hr immersing of all samples in a 3% CuSO₄, 10% H₂SO₄, 87% H₂O boiling solution (Strauss Test), the sample soaked in

sodium disintegrated. Parts of all samples were recovered, and a rate loss of weight was established for all samples exposed to the Strauss test as follows:

<u>Sample</u>	<u>% wt loss/hr</u>
As-received tube	<0.1
Argon-exposed tube--1200°F	<0.8
Air-exposed tube--1200°F	<0.9
Sodium-exposed tube--1200°F	<3.9

In a second test of a sodium-soaked sample for only 1.0-hr exposure to the Strauss test, the weight loss for one hour was 4%.

One can conclude from these screening tests that the sensitization of 304 stainless steel, by immersion in 1200°F sodium, is greater than a sample sensitized in a gas.

II. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. Gas Gain of the Proportional Counter

A number of 4π -recoil proportional counters have been utilized to measure fast-neutron spectra ranging in energy from below 1 keV to 1 MeV.⁵ In order to cover such a broad energy range it is necessary to be able to calibrate reliably the gas gain, A , of a proportional counter. This calibration can be obtained by detecting monoenergetic protons resulting from the exothergic $N^{14}(n, p)C^{14}$ reaction in the region of low gas gain. For the higher values of gas gain, the average value of the single electron response distribution was used to define A .

Fig. 25. The Diethorn Representation of Gas Gain A as a Function of Anode Voltage V

The formula of Diethorn⁶ has been used to treat the experimental gas-gain data. The gas amplification A and the anode voltage V are related by

$$\frac{\log A}{V} = C_1 \log V + C_2,$$

where C_1 and C_2 are constants characteristic of the construction and gas filling of the proportional counter. Figure 25 presents data obtained with a

counter filled to a pressure of one atmosphere of methane. It can be seen that the description of Diethorn is adequate except in the region of very high and very low values of A . At the highest values of A , the experimental results are lower than predicted, and effect attributed to saturation in the electron avalanche due to space-charge limitations in the neighborhood of the anode wire. At the low values of A , the collection mode of counter operation is no longer negligible, and this accounts for the deviation between the experimental and calculated values.

2. Fluctuations in the Fission Cross Section of U^{235}

An effort to verify reported⁷ fluctuations in the fast-neutron-induced fission cross section of U^{235} in the energy interval from 0.1 to 2 MeV was made. Both the elastic scattering and fission cross sections of U^{235} were re-examined in this region.

⁵Bennett, E. F., ANL-6897 (Sept 1964).

⁶Diethorn, W., NYO-6628 (1956).

⁷Albert, R. D., Physical Review Letters 15, 637 (1965).

Measurements of differential scattering cross sections were made by fast time-of-flight techniques at 10-keV intervals and at 8 angles between 25 and 150° with incident-neutron resolutions of 20 keV and scattered-neutron resolutions of 25 keV. The measured values were normalized to the known differential scattering cross sections of carbon, corrected for multiple scattering, and expressed as a Legendre expansion of the form

$$\frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi} \left[1 + \sum_{i=1}^4 \omega_i P_i \right], \quad (1)$$

where σ (the elastic cross section) and ω_i were deduced from the experiment. The experimental cross sections, in the format of Eq. (1), are given in Fig. 26. The cross sections of Fig. 26 vary by only a few percent from a smooth curve drawn through the measured values, and the variations were well within the relative experimental uncertainties. The coefficients ω_i also had a similar smooth energy dependence with the possible exception of one value of ω_2 , which did vary by more than a standard deviation from a quadratic least-squares fit to the measurements. However, this and other variations indicated in Fig. 26 were much smaller than the reported variation of fission cross section.

Fig. 26. Differential Elastic Scattering Cross Sections of U^{235}

The relative fission cross section of U^{235} in the region from 0.36 to 0.65 MeV were made with an automated facility that included digital-computer control of a Van de Graaff accelerator. The facility was programmed to determine the ratio of the response of a U^{235} fission counter to that of a long counter for a given statistical uncertainty and at specified neutron energies. The incident resolution was ≤ 15 keV and measurements were programmed for

5-keV intervals throughout the incident energy range. The U^{235} fission cross sections determined decreased slowly with increasing neutron energy in accord with the known general energy dependence, but specifically no structure of the type reported in Ref. 7 was evident.

3. The Fission-neutron Spectrum of Cf^{252}

The fission-neutron spectrum of Cf^{252} has been measured using a Li^6 glass scintillator as a neutron detector in combination with time-of-flight

techniques. The results over flight paths of 18.6, 36.7, and 134.2 cm are shown in Fig. 27. The solid line is given by

$$N(E)/E^{1/2} \sim \exp[-E/1.501],$$

which was obtained by a least-squares fit of all points with $0.1 < E < 10.0$ MeV. A least-squares fit of

$$N(E) \exp[E/1.501] \sim E^n$$

to all data with $E < 0.1$ MeV gave $n = 0.54 \pm 0.08$.

Fig. 27. The Fission-neutron Spectrum of Cf^{252}

B. Theoretical Reactor Physics

1. The Argonne Reactor Computation (ARC) System

The schematic diagram System shown in Fig. 28 displays the present concept for organizing the ARC System. It differs from that shown previously (see Progress Report for December 1965, ANL-7132, Fig. 15, p. 34) in that the Neutronics Preparation Module has been broken up into several modules. This modification is made necessary because code-dependent quantities are needed in Neutronics Modules and they are best computed in modules determined by the type of neutronics computation to follow.

The collection and organization of the glossary of the computational module interface quantities has been completed. The glossary is consistent with the hierarchy levels of the DATAPOOL (see ANL-7132, pp. 35-36). A typical item in the glossary contains such information as the name of the quantity, the hierarchy level in the DATAPOOL to which the quantity belongs, the definition of the quantity in terms of some basic quantities, and its FORTRAN name.

The first phase of the System will be implemented on the CDC-3600-3800. The following considerations apply:

- (a) There will be only a very limited amount of random-access storage available. Therefore, the System will make extensive use of the tapes for mass storage.
- (b) The use of tapes for the mass-storage medium, because of its nonrandom accessibility, will make the DATAPOOL organization nonoptimum.
- (c) Although the System will make extensive use of tapes, as far as possible the organization will be planned for the use of disks for large random-access storage so as to facilitate the transition (in about two years) to the CDC-6800 type of machine.

2. Calculation of Doppler Line-shape Functions

A FORTRAN program previously written to compute Doppler-broadened line shapes by interpolation of a table of the complex probability integral, based on independent six-point interpolations to find the real and imaginary parts, turned out to be slower than the older method of computing shapes by means of continued fractions. Although the FORTRAN program greatly reduced the number of arithmetic operations, inefficient compilation nullified this advantage. By rewriting the program in COMPASS, the CDC-3600 assembly language, full advantage of the simpler computational method was realized and a reduction obtained in the computation time by a factor of about three over the continued-fraction method.

This method also has considerable practical value for computing resonance cross sections. For a particular problem using an advanced version of the MC² code, the running time on the CDC-3600 was reduced from 45 to 10 min.

Fig. 28. Schematic Representation of the Present Concept of the ARC System

A set of new interpolation formulas has given improved accuracy over the older five-point difference formula discussed (see Progress Report for May 1965, ANL-7046, pp. 35-36). However, for the w function, for which derivatives are readily computed, the Taylor-series expansion is slightly more effective than interpolation.

A preliminary investigation of polynomials which deviate least from zero on a square in the complex plane indicates that it may be possible to reduce the error of the truncated Taylor series by an additional factor of about two, with no additional cost except in preparing the machine table.

C. High-temperature Materials Studies

1. Ceramics

a. Uranium-Plutonium Monosulfide Solid Solutions. The gas-metal reaction⁸ was used to prepare solid solutions of (U, Pu)S by two techniques. Alloys of uranium with 20, 40, 60, and 80 a/o of plutonium, prepared by arc-melting, were used as a starting material. Some difficulty was observed in obtaining a homogeneous solid-solution by this technique because the uranium reacts preferentially with H_2S . X-ray diffraction analysis of the final products indicated that long heat-treatments at temperatures below $1700^\circ C$ are needed to obtain equilibrium. If higher temperatures are used for equilibration, some sulfur is lost through decomposition of the unstable higher-sulfide uranium phases.

More recent work has indicated that the solid solutions are best prepared by reaction sintering of mixed powders of US and PuS. Compositions of US with 20, 40, 60, and 80 a/o PuS are being examined by metallographic and X-ray diffraction techniques. Precision measurements of lattice constants have been made for the following types of compositions: (i) pure solid solutions consisting of the (U, Pu)S phase only, (ii) (U, Pu)S in the presence of the sesquisulfide phase, (iii) phase mixtures annealed and quenched from 1900 and $1700^\circ C$, and (iv) (U, Pu)S in equilibrium with the metal phase.

The melting points of PuS and US containing less than 0.05 w/o impurities of oxygen and nitrogen were found to be $2350 \pm 30^\circ C$ and $2470 \pm 15^\circ C$, respectively. The value for US is in excellent agreement with the results of Cater *et al.*,⁹ who obtained a melting point of $2462 \pm 30 - 5^\circ C$. Compositions with sulfur contents greater than the stoichiometric amount had slightly lower melting points. The same behavior has been observed for material

⁸Annual Report for 1963, Metallurgy Division, ANL-6868, p. 140.

⁹Cater, E. D., Gilles, P. W., and Thorn, R. J., Uranium Monosulfide. I. Vaporization, Thermodynamics and Phase Behavior, J. Chem. Phys. 35, 608-618 (1961).

contaminated with oxygen; therefore, all work on the (U, Pu)S solutions will be done on high-purity material.

b. Uranium Monosulfide Oxidation. The gas-chromatograph apparatus used in the oxidation studies was rebuilt to make sample changing more convenient and to allow operation at higher temperatures. The new furnace has a $\text{Pt}_{60}\text{Rh}_{40}$ winding, which should permit the furnace to be run at temperatures of 1600°C or above. The apparatus is being recalibrated.

c. (Th-U-Pu) Phosphide Bodies. Studies have begun on the high-temperature compatibility of UP with the following refractory metals: Ti, Zr, Nb, Ta, Mo, and W. Uranium monophosphide powder was blended with 50 m/o of each of the metal powders and ball-milled for 5 hr. Pellets were fabricated with stearic acid as a binder and fired in vacuum for 2 hr at temperatures ranging from 1600 to 2200°C.

Titanium showed moderate reaction with UP at 1600°C, but the sample melted completely at 1800°C. X-ray analysis revealed the presence of a UP phase, uranium metal, and a weak unidentified phase. The lattice constant of the UP phase was 5.54 Å, compared with 5.585 Å for unalloyed UP, indicating that titanium had entered into solid solution and displaced uranium from the structure. The unidentified phase was presumed to be a titanium phosphide compound, but it was not TiP.

Zirconium reacted slightly with UP at 1600°C, but the pellet melted at 1800°C. X-ray diffraction revealed considerable solid solution, since the UP phase had a lattice constant of 5.52 Å. Uranium metal and an unidentified phase were also observed.

Niobium reacted only moderately with UP at 1800°C; however, the pellet showed signs of incipient melting and exhibited moderate vaporization at 2000°C. An unidentified phase was observed in addition to UP, but there was no sign of niobium metal in this phase. The unknown phase did not yield a diffraction pattern similar to that of NbP. The relatively large lattice constant of UP (5.585 Å) indicated that little niobium had entered the UP structure.

Tantalum showed only slight reaction at 2000°C, but the pellet melted catastrophically at 2200°C. X-ray diffraction showed unalloyed UP, tantalum metal, and an unidentified phase.

Pellets containing molybdenum and tungsten did not react at 1800°C. The composition UP-Mo showed signs of melting and vaporized considerably at 2000°C. X-ray analysis revealed an unidentified phase along with unalloyed UP and molybdenum.

The composition UP-W did not react at 2200°C. The observed lattice constant of the UP indicated that little or no tungsten had entered the rock-salt structure.

d. Thermal Stability of Plutonium Ceramics. Testing of a glovebox thermogravimetric analysis apparatus constructed for experiments with plutonium compounds continued. Temperature calibration by measurement of the apparent melting temperatures of several refractory metals, is in progress.

e. Uranium Mixed Anion Systems. An unresolved problem in the investigation of the system UC-UP has been the inability to obtain an equilibrium unit cell size for the limited solid solution UP(C). Metallographic examinations indicate a solubility of approximately 6 w/o UC at 1600°C. However, as the amount of UC increases, the unit cell size of the solid solution decreases.

Specimens of UP-10 w/o UC and UP-40 w/o UC, equilibrated at 1350°C, were refired at 1600°C, half as fragments and half as powder. The unit cell size of the UP(C) solid solution in the UP-40 w/o UC did not change appreciably for the refired fragments; however, for the powder the unit cell size increased. There was hardly any change in the unit cell size in either instance for the UP-10 w/o UC.

Samples from each of the equilibrations were analyzed for U, P, C, and O₂. The procedure for the analysis for U and P includes a step in which the material is dissolved in nitric acid. The step resulted in a light-color residue for all samples that were equilibrated as massive fragments; there was no residue from the equilibrated powders. Spectrographic analysis of the residues indicated the presence of U and P with only traces of other elements. This would suggest the presence of a phosphate, most likely a pyrophosphate because of the light color. More of this residue is being prepared for X-ray analysis. If this material is a phosphate and is proved to affect the unit cell size of the solid solution, the reactions taking place during equilibration to produce this effect are most likely complex.

2. Irradiation Testing

a. Irradiations of High-temperature Materials. A series of irradiations of ceramic fuel materials are in progress. The materials under irradiation, in instrumented capsules in the MTR, presently include carbides and sulfides. A summary of the irradiations in progress is shown in Table XIII.

A vibratorily compacted UC-20 w/o PuC specimen for nominal burnup level of 5 a/o at a jacket surface temperature of 1000°C was inserted into the MTR at the beginning of cycle 237 on January 10, 1966. Thermocouples indicated that the specimen surface temperatures were 860°C.

TABLE XIII. Status of High-temperature Fuel Irradiations in Progress

Specimen No.	Fuel Composition (w/o)	Clad Composition (w/o)	Clad ID (mm)	Thickness (mm)	Maximum Clad Surface Temp (°C)	Calculated Burnup to Date	
						a/o	f/cm ³ x 10 ⁻²⁰
MV-2	UC-20 PuC	Nb-1 Zr	6.5	0.30	660	4.4	13
MV-3	UC-20 PuC	Nb-1 Zr	6.5	0.30	760	4.9	18
MV-5	UC-20 PuC	Nb-1 Zr	6.5	0.30	750	4.7	17
MV-6	UC-20 PuC	Nb-1 Zr	6.5	0.30	670	4.9	16
C-73	UC-20 PuC	Nb-1 Zr	4.0	0.23	860	0.21	0.4
Z-1	UC-20 PuC	Nb-1 Zr	4.0	0.23	860	0.21	0.4
Z-3	UC-20 PuC	Nb-1 Zr	4.0	0.23	860	0.21	0.4
C-42	PuC	Nb-1 Zr	4.0	0.23	860	0.20	0.4
S-7	US	Nb-1 Zr	6.5	0.30	540	3.3	7.8
S-8	US	Nb-1 Zr	6.5	0.30	700	5.2	12
S-9	US	Nb-1 Zr	6.5	0.30	710	5.2	12
S-10	US	Nb-1 Zr	6.5	0.30	670	5.2	12
S-15	US	Nb-1 Zr	6.5	0.30	570	2.4	4.3
S-16	US	Nb-1 Zr	6.5	0.30	700	4.2	8.7
S-17	US	Nb-1 Zr	6.5	0.30	680	3.0	6.1
S-18	US	Nb-1 Zr	6.5	0.30	790	4.7	11

b. Fast-neutron Irradiation of Cladding Materials. The effect of elevated temperature coupled with fast-neutron irradiation in EBR-II on the mechanical properties of promising fast reactor cladding materials is being determined in the exposure range from 1×10^{21} to 1×10^{23} n/cm² and the temperature range from 550 to 750°C. The materials under investigation are AISI Type 304 stainless steel, Hastelloy-X, Inconel 625, vanadium-20 w/o titanium, and vanadium-15 w/o titanium-7.5 w/o chromium. The change in tensile, creep, creep-rupture, and tube-rupture properties of the alloys will be determined at postirradiation temperatures of 550 to 750°C.

A total of 12 capsules are being irradiated in EBR-II in two subassemblies. One subassembly contains three capsules with 16 creep-rupture specimens each of Type 304 stainless steel, Hastelloy-X, and vanadium-20 w/o titanium. They are being irradiated at calculated temperatures between 540 and 570°C, and have accumulated an exposure of 6.6×10^{21} n/cm².

The second subassembly has five capsules with tube-burst specimens and four capsules with creep-rupture specimens. Of the tube burst specimens, two capsules contain 12 Hastelloy-X specimens, two have 12 vanadium-20 w/o titanium, and one has Type 304 stainless steel. The creep-rupture specimens in three capsules are the same as in the first subassembly plus a fourth capsule containing vanadium-20 w/o titanium creep-rupture specimens. The calculated temperatures range from 540 to 570°C. These specimens have accumulated 3.3×10^{21} n/cm².

3. Liquid-metal Corrosion

a. Polarization Studies. Exploratory attempts to investigate the possibility of protecting zirconium at 400°C from corrosion in sodium by prefilming and by polarization during exposure have been completed. The temperature and/or procedure are unsuitable for the purposes of the program, because the corrosive attack on zirconium at 400°C is slight regardless of treatment. When zirconium electrodes prefilmed overnight in oxygenated sodium at 580°C (see Progress Report for October 1965, ANL-7115, p. 55) were exposed at 400°C, additional weight changes were less than $20 \mu\text{g}/\text{cm}^2$ in exposures of up to one week, well within the range of variation due to handling and weighing error. No effect of anodic polarization on surface capacitance of the order apparent in earlier tests at 560°C was observed in the 400°C tests. However, recently improved prefilming procedures (based upon dehydrogenation of the sodium and carefully controlled inert-atmosphere-glovebox techniques) yielded improved reproducibility in weight gain and surface capacitance values as well as improved appearances of the surface film. These results are otherwise consistent with previous findings.

Studies to improve polarization experiments at a sodium temperature of 540°C continue. Methods to maintain an insulating film on the portion of the zirconium sample electrode that is exposed to the sodium-vapor-phase (and argon cover gas) region of the polarization cell are being investigated. When preformed corrosion films have been employed for this purpose, their insulating character has been lost as oxygen from the film was absorbed by underlying metal at temperatures near 520°C, the sodium surface temperature. Recent approaches tested (separately) include (i) use of sulfide film on electrode shank, (ii) use of BaO_2 vapor to supply make-up oxygen to zirconium surfaces in the gas-phase region of the cell, and (iii) near saturation with oxygen of surface metal lying under film subsequently formed on the electrode shank. The first two approaches were not satisfactory. The latter method, in which oxide films of controlled thickness are formed and absorbed at 900°C, has been applied, and the results are being evaluated.

b. Dissolution Kinetics. Problems encountered in the measurements of the diffusion coefficient of Ta^{182} in liquid tin (see Progress Report for August 1965, ANL-7090, p. 39) above 800°C by the capillary-reservoir method have made it necessary to employ larger-bore Vycor capillaries. A large number of the 1.5-mm-bore capillaries used in the initial measurements exhibited breaks in the liquid column upon lowering the capillaries into the tin reservoir. These breaks were probably caused by capillarity.

In order to overcome this problem, two groups of Vycor capillaries with 2.0- and 2.5-mm-bore sizes were filled with liquid tin having Ta^{182} in solution. Ta^{182} was introduced into a tin reservoir by equilibrating

the liquid tin with a highly irradiated tantalum wire at a series of temperatures between 700 and 1000°C. As expected, the temperature dependence of the capillary activity corresponded to the temperature dependence of the solubility of tantalum in liquid tin. The capillaries filled at the various temperatures are diffused at the filling temperature or higher to insure that all of the activity is in solution. The diffusion measurements are now in progress.

Two additional dissolution experiments at 1100°C have been made at 40.0 and 180 rpm (Reynolds numbers of 2.2×10^3 and 1.0×10^5 , respectively) to explore the velocity dependence of the dissolution rate at the higher temperature.

c. Corrosion Inhibition by Dissolved Getters in Liquid-sodium Environment. The isothermal sodium solution flow loop (see Progress Report for January 1965; ANL-7003, p. 16) has been successfully tested with water (at room temperature) substituted for sodium. The novel pumping method thus having been demonstrated, the components necessary for high-temperature operation are being assembled to the basic loop structure.

The flow loop is shown to approximate scale in Fig. 29. The design provides for sodium pumping at a rate of up to 1.5 liter/min in one-inch schedule 40 pipe, yielding a flow velocity of 4.5 cm/sec in an unrestricted section of the pipe. The total volume of liquid (shown gray in the figure) contained is about 850 cc. The pump consists of two, series, in-line, ball-check valves operated at test temperature, with argon-filled void spaced above the liquid on either side. The moderate argon pressures (near atmospheric pressure) are cycled by means of bellows (operated at room temperature) driven by an electric motor through suitable mechanical linkage to effect liquid pumping. The furnaces and lower half of piping are shown in section to reveal the circulation mechanism.

Fig. 29. Isothermal Sodium Flow Loop

Furnace section A contains the sample section of the loop (left) and check-valve section (right). Samples of reactor materials under study may be distributed vertically on a holder extending down from the

upper closure. Section B contains the precipitate trap, normally to be held at the same temperature as A. Section C contains the sodium-vapor trap region and is to be operated at relatively low temperature. Section D, to be operated at room temperature, contains two stainless steel bellows of about two-liter volume each, which are alternately deflected over a 12.7-mm (adjustable) stroke fifteen times per minute.

Loop-piping material, including ball-check valves, is commercially pure nickel. This choice was based on experience in static tests. Some dissolution of nickel is expected and will be tolerated in the initial work. Static corrosion-test exposure of commercially pure nickel to sodium containing 1 w/o calcium getter for 5.5 days at 680°C resulted in small average weight changes of the order of 0.1 mg/cm² and no serious effect on appearance. Resistance of nickel to air oxidation has been observed to be satisfactory in the temperature range of interest. Although the mechanical properties appear poor at these temperatures, the requirement for strength is moderate in this application.

The advantage of the design are: (i) isothermal operation is possible near 650°C; (ii) the number of extraneous dissolved metals from the apparatus is essentially one (Ni); (iii) the quantity of impurities derived from the cover gas, through continuous reuse of a discrete quantity of argon, is limited; (iv) flow rate, addition of oxygen impurity at controlled rates and positions, and sampling the flowing liquid may be varied; (v) operation of the vessel at low pressure minimizes hazards.

D. Other Reactor Fuels and Materials Development

1. Ultrasonic Instrument and Transducer Development

Efforts to discover a suitable backing material for ultrasonic transducer probes that contain ceramic piezoelectric materials have continued.

Three sample discs of sintered Type 316 stainless steel were evaluated, with the following results:

- (a) Step-graded sample (47 to 80% dense)-- reflections from the boundaries produced high background noise.
- (b) Fine-particle sample (54% dense)-- acoustic impedance (Z) value of $13.2 \times 10^6 \text{ kg/m}^2\text{-sec}$.
- (c) Coarse-particle sample (82% dense)-- acoustic impedance value of $20.9 \times 10^6 \text{ kg/m}^2\text{-sec}$.

Low attenuation was observed in the 82% dense sample and high attenuation was noted in the 54% dense sample. From these results, it is apparent that samples would have to be continuously rather than step-graded and that fine-particle samples of higher density offer more promise than coarse-particle samples with high densities. Accordingly, fine-particle samples with densities of 65% and 75% have been ordered, as well as two sintered nickel samples.

Additional samples of tungsten-loaded epoxy were fabricated this month in an effort to obtain higher values of acoustic impedance in this material. At a loading of 13 parts by weight tungsten to 1 part by weight epoxy the acoustic impedance was about $13 \times 10^6 \text{ kg/m}^2\text{-sec}$. It appears that a loading of 13:1 is about as high as one can achieve with these materials. Higher "Z" values could probably be achieved with finer tungsten powders.

2. Development of a Neutron-image-intensification System

A second postirradiation annealing study of an irradiated fuel pin is to be made. The furnace has been prepared in such a manner that the distance between the fuel pin and the image-intensifier tube will be less than that used in the first attempt, so that improved image sharpness of the television presentation is expected. High-contrast resolution tests through the furnace indicate that a resolution capability of at least 0.75 to 1 mm can be achieved at the fuel-pin location.

In the setting up of the neutron television system, the intensifier tube was moved farther away from the reactor face than is normally the case, in order to allow room for both the furnace and the gamma shielding required during testing of the radioactive fuel pin. A significant degradation

in the resolution and contrast capability of the neutron television system was observed under these conditions. It is not clear, at the moment, whether this is the result of divergence of the neutron beam after the beam leaves the collimator, or an increase in scattered radiation.

3. Infrared Systems for Nondestructive Testing

A test plate, 1.27 mm thick, with three 0.79-mm-diameter round voids, and 0.25 mm, 0.2 mm, and 0.127 mm deep machined in the back (un-scanned) surface, was manufactured to investigate the effects of changing the transverse dimension of the void areas. All fabricated voids could be detected by the thermal detector. However, the detection of the smaller transverse voids was hindered by the introduction of erroneous signals. These erroneous signals were due to the influence of variations of the distance test sample to detector, and to room-temperature air currents.

The most significant instrumentation problem at this time is to secure a more stable detector system. Plans involve incorporation of the thermistor into a radiometer system. The detector will be capsulated, shielding it from air currents; the use of an infrared optical system for focusing should reduce the influence of detector to test plate distance variations.

4. Thermal Conductivity of Irradiated Fuel as a Function of Temperature and Burnup

Room-temperature thermal diffusivity and conductivity measurements have been completed on about half of the total number of irradiated uranium-5 w/o Fs specimens (see Progress Report for November 1965, ANL-7122, p. 54). Although all specimens from any one pin have not been tested yet, certain trends are evident. There appears to be no change in the thermal properties of the fuel pin with a burnup of 0.32 a/o. There was a substantial change in both the thermal diffusivity and the thermal conductivity for those pins with burnups of 1.8 a/o and 3.0 a/o. One specimen whose central area was marked by a large amount of "porosity" underwent about a 50% loss in thermal diffusivity and conductivity.

Preparation of the remainder of the specimens is now in progress, and they should be received within three weeks.

E. Engineering Development

1. Boiling Liquid-metal Technology

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, a pressure of approximately eight

atmospheres. Among the variables to be investigated are: Boiling heat flux and temperature difference up to the critical heat flux occurrence, boiling and adiabatic two-phase pressure losses, vapor volume fraction, and boiling stability parameters.

Modifications of the vacuum-chamber facility are continuing preparatory to installation of the Nb-1% Zr loop. The power-feedthrough and the sodium-system flanges have been completed and installed. The bus-bar connections have been fabricated. The inflatable plastic clean room has been received and installed over the chamber base to provide a clean working area over the open chamber. Ample filtered inflating air is provided by the EM pump cooling fan.

Eleven (of 55 thermocouple assemblies have been received and accepted from the vendor; the assemblies have been tested by three thermal cycles from 500 to 1850°F, and have passed a radiographic examination and a resistance test ($>10^9$ ohms from sheath to wire material). Calibration will begin immediately.

The surplus flexowriter has been modified to be compatible with the code used in the data-acquisition system and for the input to the CDC-160A computer. The flexowriter will be used to monitor the data in D-308 prior to data analysis by the computer in D-208.

The valve gaskets in the sodium-handling system have been replaced with a metal profile type and have been leak-checked. Controllers and heaters are being installed; following this, the system will be charged with sodium and operation of the zirconium hot trap will begin.

Completion of the Nb-1% Zr loop assembly has been delayed by late delivery of Nb-1% Zr foil required for finishing the double wrap of the loop tubing. The loop heaters for the preheater and boiler sections have been installed, after slight modification of the ceramic parts required for proper spacing of the radiation shields. A support bracket for the EM-pump duct has been fabricated to facilitate the transfer of the loop from the clean room to its proper position in the vacuum chamber.

b. Heater Experiments

(i) Electron-bombardment Heater Experiment. This experiment is designed to gain experience with electron-bombardment heating and to investigate geometric and mechanical factors involved in the construction of a high-heat-flux heater. The present design incorporates a sodium pool boiler with a 16-in. heated length. Although the information obtained from this experiment is being used primarily in the design of a high-temperature, high-heat-flux electron-bombardment heater, basic information pertinent to the pool boiling of sodium from a vertical cylinder may also be gleaned.

Operation of this experiment along with necessary modifications has yielded useful information pertinent to the design of an electron-bombardment heater for the Nb-1% Zr loop. Three electron bombardment heaters of different geometry have been proposed. These heaters use interchangeable components and each is designed for a different maximum heat flux. Selection of the proper electron-bombardment heater is dependent upon the experimental results and a consideration of the possibilities of installing a transient electron heater for studies of transient boiling in a forced convection loop.

Operation of the small prototype experiment continues. Modifications to the vacuum, system cleanliness, and heater geometry have extended the heater operating periods; these modifications are continually being made to examine all the variables which effect heater output and lifetime. Extensive modification to the vacuum system should permit higher vacuums during heater operation. All heater components have been cleaned and assembled, the system has been degassed, and operation is in progress.

2. General Heat Transfer

a. Heat Transfer in Double-pipe Heat Exchangers

(i) Countercurrent Turbulent Liquid-metal Flow. The practical significance of the counterflow research study was illustrated (see Progress Report for December 1965, ANL-7132, pp. 43-44) by showing the percent errors when using customary design in predicted heat-exchanger lengths corresponding to specified heat-exchanger efficiencies. Although the procedure there used could serve as an accurate method for the design of counterflow liquid-metal heat exchangers, much simpler procedures are desirable for engineering use. A simpler procedure, based on an extension of the customary design relationships, involves the use of two quantities: ϕ , called the "effectiveness coefficient"; and η , the ratio of the actual fully developed overall heat transfer coefficient to the coefficient based on the use of "film" heat transfer coefficients appropriate to the boundary condition of uniform heat flux. For a concentric tube exchanger with a "narrow" annular space, the quantity η depends on the dimensionless parameters H , K , and K_w (see ANL-7132, p. 43). The "effectiveness coefficient" depends on the same parameters as well as on the heat-exchanger efficiency (or, equivalently, the heat-exchanger length). The practicality of this "simpler" procedure depends heavily upon simplifying the dependence of ϕ on the heat-exchanger efficiency. For cocurrentflow, it was found that ϕ becomes independent of efficiency in regions of practical interest. During the past month, additional computations have revealed similar encouraging results for the counterflow case.

Improvements of the existing mercury-flow loop and the design of suitable experimental double-pipe heat exchangers continue. One of the more important purposes of these experiments will be to determine values of ϕ and η from the data obtained in ranges of practical interest, and in order to accomplish this purpose, careful design of test heat exchangers is required.

3. Electric Master-Slave Manipulator, Mark E4

The slave arm of this experimental manipulator (see Progress Report for September 1965, ANL-7105, p. 50) will be initially assembled and tested without brakes. All parts for the slave arm are on hand and preliminary tests will begin in February as soon as assembly is completed.

The design of the master arm is nearly complete; ninety percent of the drawings having been released for procurement or fabrication. Seventy-five percent of all parts are on hand; the balance is expected early in March. The master arm is slated to be assembled and the complete manipulator ready for testing by the end of March.

F. Chemical Separations

1. Fluidization and Volatility Separations Processes

a. Recovery of Uranium and Plutonium from Low-enrichment Fuels: Laboratory Support Work

(i) Fluorination of UO_2 - PuO_2 -Fission Product Pellets. A series of three runs was made with Type-II pellets¹⁰ in a 2-in.-dia fluid-bed reactor to determine the reproducibility of the experimental results under identical operating conditions. In each run a 2-in.-deep fluidized-packed bed of pellets (650 g) was fluorinated according to the sequence: (1) a two-zone oxidation-fluorination step for 3 hr at 450°C, (2) a single-zone fluorination step for 1 hr at 450°C with 10 v/o fluorine, and (3) a recycle-fluorination step with 90 v/o fluorine for 1 hr at 450°C, 3 hr at 500°C, and 8 hr at 550°C. No cesium fluoride was added to the alumina fluid-bed material in these runs. The residual concentrations of uranium and plutonium on the alumina bed for the three runs were: 0.010 w/o U, 0.010 w/o Pu; 0.006 w/o U, 0.006 w/o Pu; and 0.004 w/o U, 0.008 w/o Pu. These results from the replicate runs indicate that satisfactory reproducibility can be achieved in

¹⁰Type-II pellets were prepared by blending mixed UO_2 - PuO_2 powders and fission product oxides, and sintering the mixture in 6 v/o hydrogen in nitrogen for 6 to 8 hr at 1700°C. These pellets contain nominally 0.4 w/o plutonium, 86 w/o uranium, and 1 w/o fission products.

equipment of this type. The residual uranium and plutonium concentrations on the alumina bed obtained are similar to those obtained with Type-I pellets¹¹ under identical fluorination conditions: 0.004 w/o U, 0.007 w/o Pu.

Thus, both types of pellets behave identically toward fluorine for the removal of uranium and plutonium as hexafluorides, contrary to earlier observations (see Progress Report for November 1965, ANL-7122, pp. 59-61). In a run performed with Type-II pellets under identical fluorination conditions and in which cesium fluoride was added to the alumina bed in the amount expected from fission product cesium, the residual uranium and plutonium concentrations in the alumina bed were 0.024 w/o and 0.014 w/o, respectively. These results indicate that differences between Type-I and Type-II pellets have no significant effect on uranium and plutonium retention on the alumina bed, but the addition of cesium fluoride to the system leads to increased retention of both uranium and plutonium. Experimental investigations are underway to define the nature of the retention of plutonium by cesium and to evaluate techniques for increasing the extent of plutonium removal from systems containing cesium.

Another series of three runs was performed in the 2-in.-dia fluid-bed reactor with a 2-in.-deep fluidized-packed bed of Type-II pellets (with cesium fluoride added to the alumina bed) to demonstrate the use of BrF_5 as a selective fluorinating agent for the conversion of uranium to UF_6 . In this process, the pellet fuel is pulverized by reaction with oxygen and the powdered fuel is fluorinated with BrF_5 to form volatile UF_6 and nonvolatile PuF_4 . The plutonium is recovered as PuF_6 in a subsequent step by a high-temperature reaction with fluorine. In these runs, the pellets were oxidized with 20 v/o oxygen in nitrogen for 4 hr at 450°C and fluorinated for 1.7 to 2.0 hr at 300°C with 5 to 10 v/o BrF_5 in nitrogen. The conditions for the volatilization of plutonium by recycle-fluorination were varied among the runs to establish the conditions that would lead to high removal of plutonium from the bed. Highest plutonium removal was achieved when the recycle-fluorination step was conducted as follows: 1 hr at 300°C in which the fluorine concentration was increased from 10 v/o to 90 v/o fluorine in nitrogen, 4 hr at 300°C with 90 v/o fluorine, 5 hr with 90 v/o fluorine as the reactor temperature was increased from 300 to 550°C in increments of 10°C/12 min, followed by 3 hr at 550°C and 2 hr at 600°C. The uranium and plutonium concentrations in the alumina bed after fluorination were each 0.006 w/o. Analyses of samples of the alumina bed material taken periodically throughout the recycle-fluorination step showed that the plutonium

¹¹Type-I pellets were made from solid-solution $\text{UO}_2\text{-PuO}_2$ powder mixed with fission product oxides and sintered in 6 v/o hydrogen in nitrogen for 2 hr at 1600°C. (For further discussion of Type-I and Type-II pellets, see Progress Report for November 1965, ANL-7122, pp. 59-61.)

concentration in the alumina did not decrease during the final 2 hr of fluorination at 600°C. It appears that recycle-fluorination at this temperature is not required to achieve high removal of plutonium. Additional runs will be performed to determine if the duration of the recycle-fluorination step can be effectively shortened.

(ii) Reactions of BrF₅ with UF₄ and U₃O₈. Experimental evaluation of bromine pentafluoride as a selective fluorinating agent for uranium in uranium-plutonium mixtures continued. A series of experiments was performed in a thermobalance to determine the effects of temperature and BrF₅ partial pressure on the rates of reaction of BrF₅ with U₃O₈ and UF₄. For the UF₄-BrF₅ reaction, the temperature range was 200-275°C and the BrF₅ partial pressure was varied from 138 to 370 Torr. For the U₃O₈-BrF₅ reaction, the temperature was varied from 275 to 360°C while the BrF₅ partial pressure was varied from 89 to 369 Torr.

No effect of the linear velocity of the gas phase on the value of the reaction rate constant was observed for either reaction. The following equations were derived from the experimental data to represent the temperature and partial pressure dependence of the two reactions:

UF₄-BrF₅ reaction:

$$\log k = 0.377 \log P - 3686/T + 4.2827;$$

U₃O₈-BrF₅ reaction:

$$\log k = 0.897 \log P - 1938/T - 0.4268,$$

where k is the reaction rate constant (min⁻¹), T is the temperature (°K), and P is the partial pressure of BrF₅ (Torr).

Apparent activation energies of 16.9 and 8.8 kcal/mole were calculated for the UF₄-BrF₅ and U₃O₈-BrF₅ reactions, respectively.

b. Recovery of Uranium and Plutonium from Low-enrichment Fuels:
Engineering Work

(i) Engineering-scale Alpha Facility. The second test¹² with plutonium-containing materials has been completed in the engineering-scale alpha facility. The test involved two operations: (1) the fluorination of 8.5 kg of UO₂-PuO₂ pellets by the two-zone oxidation-fluorination process to produce a mixture of UF₆ and PuF₆, and (2) the separation of plutonium from the UF₆-PuF₆ mixture by thermal decomposition of PuF₆ to solid PuF₄ in a fluid-bed reactor.

¹²The first test and the results obtained are discussed in Progress Reports for November and December, 1965; ANL-7122, p. 63; ANL-7132, p. 47.

In preparation for the second test, the fluorinator and hexafluoride-product receiver systems were exposed to chlorine trifluoride at temperatures up to 150°C to remove moisture and oxyfluoride materials that might be present within the equipment as the result of contact with the glovebox atmosphere. The presence of moisture within the equipment would result in the hydrolysis of UF_6 and PuF_6 produced during fluorination.

The fuel charge to the fluorinator was 8.5 kg of UO_2 -0.5 w/o PuO_2 pellets. The operating conditions during the second test were essentially identical to those employed in the first test (see ANL-7122, p. 63). The alumina bed material from the first test was reused. Approximately 350 g of minus 230 mesh alumina particles were added to the bed in an attempt to reduce the holdup of plutonium-containing fines within the fluorinator (see ANL-7132, p. 47). The residual plutonium concentration in grab samples of the alumina bed after 19 hr of fluorination was 0.03 w/o, as compared with 0.025 w/o in the first test. At the end of the test essentially all the alumina fines had accumulated at the bottom of the disengaging section of the fluid-bed fluorinator. As in the first test, the fines again contained a considerable amount of plutonium. Alternative methods to reduce this holdup of fines and to return the fines to the bed during fluorination will be evaluated in subsequent tests.

A total of 9.7 kg of the UF_6 - PuF_6 product was processed through the thermal decomposer to separate and recover the plutonium as PuF_4 . The fluid-bed material for the test was minus 100-mesh refractory alumina. The mixed hexafluorides (37 v/o hexafluoride in nitrogen) were fed at a rate of 20 g/min to the thermal decomposer maintained at 295°C. The average residence time of the hexafluoride in the bed was about 10 sec. Samples of the bed material and the off-gas from the thermal decomposer have been submitted for analysis to determine the extent of plutonium removal from the mixed hexafluoride gas stream.

(ii) Process Development Studies for Uranium Dioxide Fuels.

A test was performed in the pilot-plant facility to demonstrate decladding of Zircaloy-2-clad uranium dioxide fuel bundles. The facility consisted of a 6-in.-dia fluid-bed reactor, a 9-in.-dia downflow packed-bed filter, and a 6-in.-dia pyrohydrolysis reactor for the conversion of volatile chlorides to solid oxides. The fuel bundle consisted of two solid Zircaloy-2 rods and four Zircaloy-2 tubes packed with UO_2 pellets; both the rods and tubes were 70 in. long. The fluid-bed depth was 14 in. The fuel bundle was contacted with 48 to 64 v/o hydrogen chloride in nitrogen at a bed temperature of 400°C. Decladding was completed in 4.5 hr.

After 2 hr of reaction one of the Zircaloy rods was taken out of the reactor and the diameter measured along the entire length. The penetration rates, calculated from dimensional changes, averaged 17 mils/hr

for the portion of the rod immersed in the fluid bed and 34 mils/hr at the interface between the fluid bed and the gas phase. The highest rate of reaction occurred at the portion of the rod which was located above the fluid bed in the zone where the diameter of the reactor increases from 6 in. to 12.5 in. The portion of the rod above the fluid bed was covered with a friable scale of varying thickness (2 to 10 mils). No scale was observed on the portion of the rod that had been immersed in the fluid bed. Evidently the scale had been removed by the action of the fluidized bed.

A temperature profile was obtained for the second Zircaloy rod from twelve thermocouples that were spot-welded to the rod at 6-in. intervals. It was observed that, in the fluid-bed zone, the temperature of the rod was identical to that of the fluid bed ($\sim 400^{\circ}\text{C}$), thereby indicating that the heat transfer from the surface of the rod to the fluidized bed was good. In the gas phase above the bed, the temperature of the rod was much higher, up to 640°C , although the temperatures of the surrounding gas and the reactor wall were 400°C , thus indicating that the heat transfer from the rod to the surroundings was poor. The highest temperature was observed at a position equivalent to the position where the highest penetration was measured on the companion rod.

On the basis of these data on penetration rates and temperature measurements, it probably is possible to de-clad long fuel bundles in shallow fluid beds. Experiments with larger tube bundles in larger reactors are needed to establish this point firmly.

(iii) Studies with Irradiated $\text{UO}_2\text{-PuO}_2$ Fuels. The first test with irradiated Zircaloy-clad $\text{UO}_2\text{-PuO}_2$ fuel has been completed in the bench-scale fluid-bed unit installed in the Chemical Engineering Division's senior cave. The test was temporarily discontinued when biologically significant quantities of Ru^{106} activity were found in the stack discharge from the cave to the atmosphere. Various methods for trapping ruthenium were tested; a method involving the use of a bed of sodium fluoride pellets at 400°C was found to be effective and the test was resumed. The use of sodium fluoride resulted in a decrease of the Ru^{106} concentration in the stack effluent to a level which was 10% of the allowable concentration. The total quantity of Ru^{106} discharged to the atmosphere during the test was 2.2 mCi which represented 0.03% of the Ru^{106} charged to the fluid-bed reactor in the fuel material. This quantity of ruthenium is considerably below the permissible discharge over the period of the test. Experiments are planned to evaluate other methods for the effective trapping of ruthenium and for the identification of the nature of the ruthenium in the effluent gas stream.

G. Sodium Coolant Chemistry

1. Control of Sodium Oxide Impurity

The Kiethley electrometer used with the oxygen electrochemical cells has been sent back to the factory for repair. Pending its return tests with the plugging meter have continued. Plugging runs have been made under four conditions, i.e., electromagnetic pump in plugging loop on or off; and the cooling fan blowing across the plugging valve on or off. Although the maximum effect of these variations results in an almost doubling of the time required for a plugging run, the plugging temperatures obtained are surprisingly close for the four varying conditions. In general, the oxygen content of the sodium as indicated by the plugging indicator, correlates within a maximum error of about 20% with oxygen content as obtained by amalgamation analysis.

H. Plutonium Recycle Program (EBWR)

1. Reactor Plant Preparation and Maintenance

Repairs of piping welds in the 8-in., primary high-pressure steam line leading to the EBWR reboilers as being completed. In the majority of cases, the old welds were cut out and replaced with new. Radiographic examinations of the finished welds showed them to be sound.

Extensive shrinkage casting defects in the 3-in. motorized feedwater stop valve (Pw-168) were successfully repaired by welding. After lapping the mating seat members, a 1000 psi hydrostatic test applied against the seat of the valve demonstrated the valves' integrity and suitability for further service.

A detailed stress analysis of the existing, and of an alternative, 8-in. primary piping system between the reactor vessel and the primary reboilers was completed. The computer analysis revealed that both piping schemes were suitable. Furthermore, the analysis resolved the problem of rebuilding a substantial part of the steam system inside the reboiler building. A reinforced lateral connection supplying high-pressure steam to both reboilers will be fabricated to replace the present connection, which has a cracked elliptical intersection weld (the 6-in. branch connection from the 8-in. supply line).

2. Temperature Coefficient of Reactivity

The moderator temperature coefficient for the 36-assembly loading with boric acid concentrations near 5 g/gal was remeasured by a method based on integral rod worths. At a given temperature, the boric acid

concentration was adjusted so that the reactor was critical with all control rods withdrawn except one. The exception was Rod No. 4, a corner rod, which was partially inserted. From the integral worth of the rod obtained from period measurements, the reactivity of the system is determined as a function of temperature and boric acid concentration. The results are summarized as follows:

Temperature (°F)	H ₃ BO ₃ Conc (g/gal)	k	k for an H ₃ BO ₃ Conc of 4.80 g/gal
67.1 ± 0.3	4.80 ± 0.02	1.00065 ± 0.000032	1.00065 ± 0.000032
94.1 ± 0.2	4.86 ± 0.02	1.00091 ± 0.000045	1.00166 ± 0.00032
122.5 ± 0.1	5.09 ± 0.02	1.00056 ± 0.000028	1.00418 ± 0.00042
135.5 ± 0.1	5.19 ± 0.05	1.00000 ± 0.000001	1.00487 ± 0.00050

To obtain a temperature coefficient, k is computed for a boric acid concentration of 4.80 g/gal and a boric acid worth of -0.0125 ± 0.0012 . $\Delta k/g/gal$. From these data an average moderator temperature coefficient of $(6.21 \pm 1.1) \times 10^{-5}$ between 67°F and 135°F was obtained. At the present time one-dimensional and two-dimensional, four-group diffusion theory problems are being prepared for calculation of the temperature coefficient of the plutonium critical with rods withdrawn. Unfortunately, because of the complicated structure of the control-rod followers, the experimental configuration cannot be exactly simulated in these calculations. However, the temperature coefficient of the full system is of greater interest and will be measured later.

III. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Reactor Physics Experiments

Deliveries of fuel foils have now augmented the supply of standard (0.0044-in.-thick) fuel foils to the point that specific scheduling of core unloading and reloading could be established. Experiments with the present core (810 fuel foils and 810 boron stainless steel poison strips) have been completed, and the core was unloaded.

The fuel assemblies were disassembled and the jacketed fuel foils inspected. Temporary shielding was used to reduce whole-body exposure of the personnel because of the high residual fission-product activity.

The core was loaded with a full complement of 1215 fuel foils, containing ~62.1 kg U^{235} . The number of boron-stainless steel poison strips was increased to 1620 (850 g boron). Criticality was achieved with 2 in-core control blades (Nos. 9 and 11) fully inserted, and in-core blade No. 7 very slightly inserted.

A 4-in.-thick slab of steel has been installed at a radial distance of 4 ft from the vertical axis of the Internal Thermal Column, to simulate the presence of the 8-ft-ID AARR pressure vessel. It is expected that the actual pressure vessel will be slightly thicker, but this steel slab will be useful in planned measurements of neutron and gamma-ray fluxes in the vicinity of the slab.

More accurate measurements have been made to determine the losses in available excess reactivity--and in control-blade worths--that would occur if stainless steel control-blade followers were to be used instead of, e.g., Zircaloy. (In the Criticality Facility, the normal control-blade followers are aluminum.) For in-core radial control blade No. 7, a stainless steel follower reduces the total available excess reactivity by 0.1% when the control blade is in its position of full withdrawal, as compared with the case of a full blade follower of aluminum. This means that the reactivity control capacity of the blade is reduced by 0.1%. Similar, accurate measurements for peripheral control blade No. 2 indicated a reactivity loss of slightly more than 0.3% for a stainless steel follower; previously (see Progress Report for November 1965, ANL-7122, p. 75) the loss had been estimated as ~0.4%. This loss in available excess reactivity is too large to justify the use of stainless steel followers for the peripheral control blades.

Detailed U^{235} -fission mappings in the vicinity of control-blade followers are in progress. If the power peaking near blade followers is reduced substantially by the use of stainless steel blade followers, and if

otherwise it is feasible to use such followers, the reactivity loss can be borne for the 6 in-core control blades. Preliminary data indicate significant power peaking near in-core control channels, which are thinner (0.25 in.) than the channels are expected to be in the actual AARR (0.32 in., or more).

Additional measurements have been made with sample control blades containing europia (Eu_2O_3) stabilized with titania (TiO_2) or with hafnia (HfO_2) (see ANL-7122, p. 75). The complete sets of data are presented in Tables XIV, XV, and XVI.

It should be noted that in all measurements, stainless steel followers were used. As indicated above, the use of a stainless steel follower for a peripheral control blade causes a very large reduction in control-blade worth: $\sim 0.3\%$. It is partly for this reason that the adjusted peripheral-blade worths (see Tables XV and XVI) are smaller than the worths in in-core positions, in contrast with data of normal control-blade worths in the Criticality Facility. The normal blade-follower for the Criticality Facility is aluminum, not stainless steel.

TABLE XIV. Descriptions of Control-blade Specimens^a

Specimen Abbreviation	Composition of Absorber Portion (w/o)	Dimensions of Control Blade ^b (in.)		Estimated Dimensions of Absorber Portion (in.)		
		Thickness	Width	Thickness	Width	Length
Hf	pure Hf	0.15	7.5	0.15	7.5	$26\frac{1}{4}$
S1	37% Eu_2O_3 ; 16.6% TiO_2 ; 46.6% SS	0.213-0.215	5.75	0.18	5.18	$18\frac{5}{8}$
S2	31.3% Eu_2O_3 ; 14% TiO_2 ; 54.6% SS	0.194-0.198	5.75	0.17	5.30	$18\frac{1}{2}$
S3 ^c	ZrO_2 plus 49.3% SS	0.204-0.206	5.75	0.17	5.14	$18\frac{1}{4}$
M1	32% Eu_2O_3 ; 14.3% TiO_2 ; 53.7% SS	0.209-0.211	5.59	0.18	5.35	$18\frac{5}{8}$
M2	19% Eu_2O_3 ; 22.6% HfO_2 ; 58.6% SS	0.203-0.210	5.49	0.19	5.26	18

^aIn some cases, the literal sum of weight percentages slightly exceeds 100%. These are round-off errors.

^bThe cladding is SS.

^cThis specimen is included to permit inferences about the reactivity worth of the ceramic portion of the control plate.

TABLE XV. Reactivity-control Worths of Blade Specimens
When Halfway into the Reactor Core^{a,b}

Specimen Abbreviation	Measured Reactivity-control Worth (%)		Adjusted Reactivity-control Worth ^a (%)	
	In-core Blade	Peripheral Blade	In-core Blade	Peripheral Blade
Hf	0.540	0.478	-	-
S1	0.593	0.446	0.659	0.625
S2	0.582	0.450	0.631	0.616
M1	0.625 ^c	0.460	0.672 ^c	0.645
M2	0.535	0.415	0.585	0.572

^aThe adjusted values are based on proportionality to absorber width. In-core blade position No. 8 was used, with a reference absorber width $5\frac{3}{4}$ in. Peripheral-blade position No. 4 was used, with a reference absorber width of $7\frac{1}{4}$ in.

^bThese worths were measured with stainless steel followers attached to the blade specimens.

^cNew data; note revisions of data reported in ANL-7122, p. 75.

TABLE XVI. Reactivity-control Worths of Blade Specimens
When Fully Inserted into the Reactor Core^{a,b}

Specimen Abbreviation	Measured Reactivity-control Worths (%)	
	In-core: Position No. 8	Peripheral: Position No. 4
S1	1.13	0.88
S2	1.13	0.91
S3	-0.022	-0.024
M1	1.15	0.88
M2	1.07	0.83

^aAverage of 2 measurements; values not adjusted for absorber width.

^bIn these measurements, stainless steel followers were attached to the blade specimens. Therefore, to normalize to the case of an aluminum follower, the worths of in-core control blades should be increased by +0.1%, and the worths of peripheral control blades should be increased by approximately +0.3%. Each of the control-blade specimens (excluding S3) is worth more than the blade normally used in the Criticality Facility.

In recent experiments with simulated beam tubes, the relative levels of neutron flux, and the reactivity change as a function of distance from the reactor core, have been measured. The tubes are square, in cross section, with outside dimensions 4 in. x 4 in. In Fig. 30 are shown the arrangements of the beam tubes and collimators used in these measurements. Data presented in Figs. 31 and 32 were obtained by varying

Fig. 30. Arrangements of Beam Tubes and Collimators (not to scale)

Fig. 31. Relative Count Rate in Stationary BF_3 Counter at the End of the Tangential Beam Tube vs. Position of Source Plane

Fig. 32. Relative Count Rate in Stationary BF_3 Counter at the End of the Tangential Beam vs. Position of Source Plane with 1-in.-thick Lucite Layer at Tip

the position of plugs of beryllium, or of beryllium and Lucite, in the tangential tube. In these experiments, a collimator made up of cadmium and borated paraffin was employed (see Fig. 30), and a stationary BF_3 counter was used for neutron detection.

Figure 33 summarizes results with indium detectors, using the compound collimator described above, or a simple cadmium collimator (see Fig. 30). In the measurements with indium foils, the entire tangential tube was moved, contents and all, to standardize geometry. The indium activations are believed to yield the more accurate measurements of relative "thermal" neutron flux.

Fig. 33. Relative Sub-cadmium Activations with Indium Foils at Ends of Beam Tubes

The relative normalization between curves (Figs. 31 and 32) may be erroneous since, in some respects, these curves do not match the indium-foil data. Figure 33 includes data both for the radial tube and for the tangential tube, but only for a limited range of positions.

2. Theoretical Reactor Physics

The relative effectiveness of fission-spectrum sources in determining the magnitude of the peak thermal-neutron flux in the outer beryllium reflector is shown in Fig. 34 for the reference graded-fuel core. The curves are analogous to those presented previously for the Internal Thermal Column (see Progress Report for November 1965, ANL-7122, p. 72, Fig. 21). Note that the

curves for groups 15 and 16 would almost coincide if plotted with the same reference zero on the ordinate. The curves are exponential in shape, as was the case for the analogous curves for the ITC.

Fig. 34. Relative Contribution by Fission-spectrum Sources in the Core to the Peak Thermal-neutron Flux in the Beryllium Reflector (Normalized to Unit Contribution by Source at Outer Boundary of Reactor Core)

The implications of these curves are being studied. A tentative conclusion is that it would be preferable to flatten the gross radial power distribution still further, at some sacrifice to the optimum values of the peak flux per unit reactor power available for experimenters. Computations are in progress to determine the feasibility of achieving the additional power flattening by, for example, increased grading of the U^{235} loading. Eventually the variation of the gross radial power distribution as the reactor is operated will be studied.

A computation has been made of the change in available excess reactivity if the reference fuel loading could be increased by 20%, with an appropriate reduction in the content of stainless steel in the cermet. If no boron were present, the reactivity gain would be $\sim 1.4\%$. However, in the presence of enough boron to make the reference reactor critical, a 20% increase in U^{235} loading yielded a reactivity change of 4.7%. At first consideration this result might seem to be surprising. The reason for this effect is that the reactivity-control worth of the boron, initially $\sim 16\%$, is

reduced markedly by the increased relative neutron absorption in the fuel. This reduced effective poisoning by the boron corresponds to a substantial additional gain of reactivity. The lesson to be learned from this analysis is that, inversely, as fuel is consumed during reactor operation, the poisoning worth of each gram of burnable poison (boron) remaining increases somewhat.

Calculations have been made of the neutron-flux spectrum as a function of radial position in the fuel zone and as a function of the loading of fuel and boron. Table XVII summarizes the results for various loadings in the Criticality Facility, listing the median neutron energy for the production of fission-spectrum neutrons. In condensed form, this table, for the Criticality-facility loadings, is complementary to Table XVI of the December 1965 Monthly Progress Report, ANL-7132, p. 67. Table XVIII lists this median neutron energy for the asymptotic portion of the reactor

TABLE XVII. Median Neutron Energy for Fission-neutron Production in Various AARR Criticality-facility Loadings as a Function of Radial Position in the Fuel Zone^a

Core Loading	r = 5.88 cm (Inner Edge of Core)	r = 7.10 cm	r = 14.79 cm (Center of Core)	r = 22.08 cm	r = 23.30 cm (Outer Edge of Core)
315/0	16	16	15	16	16
615/420	16	15	14	15	16
810/810	16	15	12	14	16
1215/2000	16	12	11	11	16

^aThe tabulated numbers are the identifications of the neutron-energy groups of the 16-group calculation. Neutron energy ranges are:

Group No.	Energy Range (eV)	Group No.	Energy Range (eV)
11	10.0-30.0	14	0.4-1.0
12	3.0-10.0	15	0.1-0.4
13	1.0-3.0	16	0-0.1

TABLE XVIII. Asymptotic Median Neutron Energy for Fission-neutron Production as Function of Thermal Absorption Cross Section

Core Loading	Macroscopic Absorption Cross Section in Group 16, Σ_{a16} (cm ⁻¹)	Asymptotic Median Neutron Energy, Group No.
315/0	0.40	15
615/420	0.74	14
810/810	0.97	12
1215/2000	1.53	11
Reference Fuel	1.37	11 ^a

^aGroup No. 11 covers the neutron energy range from 10 to 30 eV.

fuel zone as a function of the total macroscopic absorption cross section in neutron-energy group No. 16. Note that only the neutron energy group is listed, since the multigroup calculations are neither sufficiently detailed nor sufficiently unambiguous to determine the median neutron energy more accurately.

Safety-analysis computations have been made for the reactivity effects of flooding of beam tubes and through tubes in the reference AARR reflector. The aluminum tubes and the associated void and coolant water were assumed to be effectively homogenized with the local beryllium environment. Flooding of one tube was assumed to correspond approximately to the replacement of 10% of the total void volume in the beryllium by water. The computed result was a reactivity loss (roughly -0.01%). Flooding of 30% of the beam tubes yielded a computed reactivity loss of 0.05%. Flooding of all of the beam tubes resulted in a computed reactivity loss of 0.18%.

Earlier experiments in the Criticality Facility, with no water in the beryllium reflector, demonstrated that flooding of one tube (4 x 4 in. outside dimensions) caused a very small change in reactivity--probably positive (see Progress Report for June 1965, ANL-7071, pp. 55-56). Flooding of the two beam tubes and the through tube caused a 0.2% loss in reactivity. If there had been water in the beryllium initially, almost certainly flooding of one or any combination of beam tubes would have yielded, at worst, a small reactivity gain--probably a reactivity loss.

3. Heat Transfer

a. Experimental Studies. A study to determine the suitability of a constant-potential (150 kV) X-ray machine for making void measurements during transient heat transfer tests was started.

An existing X-ray machine together with a typical AARR test section, complete with pertinent hardware, has been used in tests with an apparatus for simulating void oscillations. The X-ray beam was passed through the test section. The output on a photomultiplier was read as void level changed. Measurements and pictures of the output on an oscilloscope were made and studied.

The results indicate that the ripple in the beam is high by a factor of about ten over that estimated to be required for the transient tests. The problem appears to be related to the regulation of the power supply for the X-ray equipment. Means of correcting the situation are being sought from X-ray equipment manufacturers.

A test loop for conducting shutdown emergency cooling tests has been constructed and has undergone successful hydrostatic tests (see Progress Reports; for December 1964, ANL-6997, p. 52, for May 1965, ANL-7046, p. 74 and for July 1965, ANL-7082, pp. 58-59).

4. Fuel and Materials Development

Investigations at Battelle Memorial Institute to establish a reference process for the addition of boron burnable poison were completed. Zirconium diboride (ZrB_2) was chosen as the reference material because of its large negative free energy. However, in view of the problem of loss of boron in fabrication of UO_2 -stainless steel cermet fuel containing ZrB_2 , special emphasis was placed on selecting a material and process for coating the burnable poison so as to reduce the loss. The coating process established as reference in these studies consists of an initial preparatory cover of 1-2 μ of chromium carbide, followed by a subsequent layer of 4-6 μ of niobium. Destructive examination of numerous fuel samples which had been fabricated under varied temperature conditions indicated that at temperatures above 1200°C a complete reaction occurred between (1) bare ZrB_2 and Type 347 stainless steel, and (2) niobium-coated ZrB_2 and Type 347 stainless steel. In both cases a eutectic had formed and distributed itself throughout the matrix grain boundaries.

At temperatures between 1175°C and 1200°C the reaction was reduced significantly by the niobium coating, and at temperatures below 1175°C very little reaction occurred in either coated or uncoated ZrB_2 . Factors other than temperature alone were observed to influence the rate of reaction, but these were controlled in the fabrication verification studies.

Since optimum plate-rolling temperatures which reduce UO_2 -particle crushing during rolling are very close to these temperatures, the Cr_3C_2 - and niobium-coating technique is being retained as the reference process; samples fabricated by this method will be used in the next set of ETR irradiation tests planned.

Continued postirradiation investigations by Phillips Petroleum Company at the ETR facilities have indicated that the highly loaded, low-density UO_2 -Type 347 stainless steel dispersions for AARR can be expected to perform as predicted according to the relationships developed by ORNL¹³ and by Keller at BMI.¹⁴ Blister tests conducted in the hot cell showed that the fuel having 23-24 a/o U^{235} burnup (i.e., $\sim 3.5 \times 10^{21}$ fissions/cc UO_2) developed blisters when heated to $\sim 950^\circ\text{F}$. Fuel having an average burnup of 17 a/o U^{235} burnup (i.e., $\sim 2.5 \times 10^{21}$ fissions/cc UO_2) developed blisters when heated to 1170°F .

Figure 35 shows a typical cross section through a blister in the 37 a/o UO_2 -347 stainless steel irradiation specimen. Examination of the

¹³Irradiation Testing of Core "B" for the Enrico Fermi Fast Breeder Reactor, ORNL-3709 (Nov 1964), p. 48.

¹⁴Donald L. Keller, Predicting Burnup of Stainless- UO_2 Cermet Fuels, Nucleonics, 19, No. 6, 45 (June 1961).

failed section showed crack propagation occurred mainly at the major diameter of the particle, and separation occurred at the area of highest density of UO_2 . It is postulated that the reason why the behavior of the 37 w/o UO_2 -stainless steel cermet agreed with that of lower w/o UO_2 -stainless steel cermets used in Keller's and ORNL's correlations is that special care was given to the provision of fission gas reservoirs in the UO_2 (in the form of reduced UO_2 density), thus reducing the net internal pressure on the matrix. Also special care was given to optimizing the homogeneity of the dispersion, thus improving the uniformity of the matrix ligaments around the particles. This attempt to reduce the pressure on the matrix from fission product buildup, and optimization of matrix ligament geometry around the particles, appeared to be sufficient to compensate for the reduction in matrix material in the 37 w/o UO_2 -stainless steel cermets (as compared to the quantity of matrix material in the 20-30 w/o UO_2 -stainless steel materials used in the references).

Fig. 35. Cross Section of Postirradiation Blister Test Specimen Showing Typical Pattern of Failure at Maximum Temperature of 950°F after Being Irradiated to $\sim 3.5 \times 10^{21}$ fiss/cc- UO_2

Although these tests are not conclusive, they do indicate that the 37 w/o UO_2 -stainless steel cermet with a nominal 85% T.D. (theoretical density) UO_2 in an optimum dispersion appears to be limited to a maximum fuel burnup of approximately 25 a/o U^{235} at the maximum fuel centerline temperatures expected in the AARR Mark-I core during 240-MW operation.

Irradiation testing of GE-8100 brazed stainless steel joints has been started in the ETR-G12. The shear strength of the joint in the unirradiated

condition is in the range from 30,000 to 60,000 psi. These tests will attempt to determine the overall change in the shear strength due to irradiation as well as to evaluate the change in ductility.

Work on a process for the fabrication of all-sintered fuel by the 3M Company has been terminated because the company has stopped its nuclear fuel activities. Prior to the termination of the development program for AARR, sufficient process-development studies were completed to enable resumption of this development work by others should the need for this type of fuel become urgent in the future. However, the previously planned production of full-size plates was not carried out, since it was believed that this work should be reserved until the need actually arises, and then should be performed by an organization which would be in a position to undertake production of reactor cores if desired.

5. Reactor Vessel

A Selection Board has been established to select, on the basis of technical competence, a vessel manufacturer to perform the detailed design of the AARR reactor vessel on a cost-plus-fixed-fee contract. The contract will also provide for the selected firm later to fabricate the vessel, either for a lump sum or on a further cost-plus-fixed-fee basis (at the option of the Laboratory). This latter option will be negotiated after completion of the Title I plant design by the architect-engineer. The preparation of the vessel design specifications and drawings, which will form a part of the contract proposal invitation package, is in progress.

6. Control Rod Drive Mechanism

Sixty tests of clutch delay time with a 10,000-ohm resistor and a 0.47- μ fd capacitor in parallel with the clutch coil resulted in an average delay time (time between power interruption and shaft movement) of 4.5 ms with a maximum discharge voltage of 900 V. The clutch manufacturer has stated that the coil insulation was rated at 3000 V dry, and 1500 V wet, to 250°C. Clutch conditions for the AARR rod drive will be normally dry. The subject clutch has been incorporated into the prototype drive design.

A 5-ft-long dummy control rod has been tested by giving it sixty full-stroke scrams at an acceleration force of 7 g. The Type 304 stainless steel rod showed no ill effects. Inspection of the connecting pins revealed a crack in the 5/16-in.-dia pin but no damage to the 7/16-in.-dia pin. The 5/16-in. pin will be replaced with one of 7/16-in. dia and the tests continued.

7. Development of Experimental Facilities

Tests to determine the effects of exposed beryllium surface area and of exposing aluminum in the same system with beryllium on the corrosion

rate of beryllium has been completed. The tests were conducted at 200°F in deionized water flowing at 44 fps. The volume of water was 27.6 liters. The following table summarizes the results.

Test Time (hr)	Exposed Surface Area of Beryllium (cm ²)	Corrosion Rate (mpy)
1150	12.5	2.7
1628	125.0	1.1
1118	12.5*	3.7

*113 cm² 6061 aluminum exposed in the same loop.

The results indicate that increasing the surface area of beryllium decreases its corrosion rate. Although the reported values indicate that the simultaneous exposure of 6061 aluminum with beryllium accelerates the corrosion rate of beryllium, it is probable that the difference between 2.7 and 3.7 mpy is not significant and only represents the limits of reproducibility. In all three tests, beryllium corroded uniformly and developed only very thin films. These results indicate that the general corrosion rate of beryllium in the AARR will be acceptably low.

A relatively long-term isothermal test is in progress to evaluate the effects of crevices and galvanic coupling on the corrosion of beryllium, aluminum, and stainless steel. The test conditions are identical to those used in the beryllium tests.

The 6061 aluminum specimen subjected to a heat flux of 400,000 Btu/hr-ft² (see Progress Report for December 1965, ANL-7132, p. 71) had been in test for 1527 hr. The greatest temperature increase observed at any thermocouple location was 12.6°F, which corresponds to a corrosion product thickness on the heat transfer surface of about 0.5 mil.

Shortly after 1527 hr of operation, a power failure caused interruption of the test. As soon as the cause of the failure is determined and corrected, it is planned to restart the test and continue it for a total of 2000 hr.

IV. NUCLEAR SAFETY

A. Reactor Kinetics

1. Fast Reactor Safety

a. Coolant (Water) Expulsion Studies. The experiments with water have been extended to larger rates of energy input. The preliminary trends have indicated that the magnitude of the initial pressure pulse increases with increasing rate of energy input and initial water temperature.

Several experiments were conducted with other liquids, notably methanol and ethylene bromide. Analysis of the data shows that the dense ethylene bromide (specific gravity ≈ 2.5) yields, for similar conditions, significantly larger pressure pulses than water.

It also has been observed that the "water-hammer" pressures (caused by the two-phase, water-steam mixture falling back in the tube) do not occur when the liquid is near its saturation point. This large pressure pulse occurs only for significantly subcooled liquid.

b. Two-phase Flow and Related Problems. A program has been initiated to measure void fractions (velocity ratios) under critical flow conditions with steam-water as the working mixture. Specifically, a detailed investigation of the fluid behavior at the point of choking in rectangular and circular constant-area ducts will be carried out. The data obtained from these experiments is required to evaluate accurately the existing critical flow models, such as those of Fauske, Levy, and Moody.

Data already obtained indicates there is no maximum in the critical flow rate as a function of quality as is predicted by both Levy and Moody. These flows are also higher than those predicted by Fauske's model.

Void-fraction measurements in the rectangular channel indicate the slip is nowhere near as large as predicted by the above models. The measurements also show sizeable metastable effects in choked flow.

2. Analysis of Meltdown Experiments

A typical hypothetical fast reactor meltdown incident is a multistage event, proceeding as follows: first, there is some operating abnormality or overall accident, which produces some degree of fuel failure; second, fuel moves rapidly into a more reactive (nuclear) configuration; third, the reactivity increase resulting from the movement causes a destructive nuclear burst. In recent years, this definition has been expanded to include such phenomena as fuel motion occurring without failure (for example, oxide fuel moving down a central void in the fuel pin) and reactivity increases resulting from coolant movement.

The principal reasons for the need to study this type of "maximum design" accident include the following: first, the typical fast power reactor core configuration is considerably less compact than the most efficient arrangement, and thus it is relatively easy to postulate motions that could cause sizeable reactivity increases; second, the prompt-neutron lifetime is so short that super-prompt-critical reactor periods are $\lesssim$ a few milliseconds--too short to permit effective control by mechanical devices; third, the mechanisms which must be understood in order to characterize the motions are sufficiently complex and little studied that extreme conservatism has been relied upon in accident analyses as compensation.

Because of this third point, an experimental program is underway, at TREAT to study the phenomena of meltdown, the mechanisms producing movements, and the interrelations concerned. There are two basic routes that can be followed: one relies mainly on sample fissions induced by thermal and near-thermal neutrons; the other uses loops shielded from low-energy neutrons. The first approach requires consideration of neutron-flux depression and self-shielding effects, which can be partially compensated in tests on clusters of pins by using pins with different enrichments in different rings. The second approach uses high sample enrichments, but has much less severe shielding problems, and produces much smaller changes in flux distributions during movement of fuel in a meltdown experiment. Because representative fast reactor fuels contain $\sim 20\%$ plutonium, experiments on clusters of such pins appear to require the nonthermal-flux approach.

TREAT has been used successfully for meltdown experiments with samples shielded by cadmium. EBR-II-type pins of Th-20 w/o U have been run well past the failure threshold in cadmium-lined transparent meltdown containers (see Progress Report for January 1965, ANL-7003, pp. 77-79). Uranium was fully-enriched so that the alloy contained about 18.6 w/o U^{235} . The energy release for these samples, based on radiochemistry, was 1.61 J/g of fuel alloy per MW-sec of TREAT energy release. (This value was in good agreement with other estimates based on postexperiment condition of the fuel and on pre-experiment calculations.) This experimental number indicates that the order of 8 kW-sec energy release per gram of U^{235} can be obtained in a cadmium-shielded sample subjected to the current maximum TREAT transient of 1000 MW-sec. This energy release from a sample is about 8 times that necessary to raise fully enriched UO_2 from room temperature to its melting point under adiabatic conditions. Alternatively, it is enough to permit running oxide fuel at the interesting power rating of 200 W/g of fuel for about 40 sec.

Because of the long thermal time constant typical of oxide fuels, it is necessary to consider whether or not internal fuel-temperature distributions typical of steady-state design conditions can be obtained in the

early stages of a TREAT transient without either overheating the lower-temperature regions (which would occur in the case of too-rapid initial heating) or expending too much of the reactor energy capacity. Transient calculations show that internal temperature distributions at steady state can be obtained in a manner consistent with TREAT kinetics characteristics for reasonable values of the sample to reactor energy ratio. Calculated maximum central fuel-temperature rises were close to those expected of an adiabatic process.

More detailed investigations into use of TREAT as a fast flux loop meltdown facility are underway, in preparation for exploitation of the reactor capability for this type of operation.

3. Containment Program

A memorandum outlining a major structural program for the safety of large fast breeder reactors is under review by the management. A detailed program of theoretical and experimental work is given along with estimates of the time, money, and personnel to accomplish the program.

An analytical study of the dynamic response of an infinitely long, thick, elastic cylindrical shell to an axisymmetric pressure pulse has been initiated. This represents the first step towards more realistic geometrics for studies of reactor safety. The main effort is involved in trying to develop integral transforms so that the problem of arbitrary loads on both surfaces can be solved.

B. TREAT

1. Operations

Two metal-water reaction experiments were performed in transparent capsules to extend reaction temperature and photographic data on the physical behavior of the samples while the reaction is in progress into the region of extensive fuel fragmentation. One of these samples was a SPERT-type uranium-aluminum alloy, and the other was a HFIR U_3O_8 -Al cermet.

Two samples of prototype PBF fuel were irradiated for the Phillips Petroleum Company. One sample was water-logged prior to a transient designed to simulate normal PBF fuel temperatures. The other sample was subjected to a transient designed to simulate the PBF fuel temperature anticipated for a 150% overpower condition.

2. Large TREAT Loop

Installation of the blanket gas system is nearly complete. The remainder of the heated systems in the sodium equipment room is being insulated.

The Zircaloy liner, graphite blocks, and upper liner section for the in-pile region have been assembled and fitted to allow proper clearances. The items which have been received from the shops in the past month include the crossover bellows assemblies for the horizontal pipe sections, the test-section pressure vessel, and insert tube. The vendor has difficulty in making the final weldments on the surge-suppressor vessel; delivery will be delayed somewhat. The test section will be checked for mating parts fit, after which instrumentation will be installed.

The EBR-II transfer lock cover overturned the transfer cask, which subsequently fell upon the TREAT loop-loading tank with resultant damage to the tank and need for extensive repairs. This incident will delay filling the out-of-pile section of the loop with sodium.

C. Chemical and Associated Energy-transfer Problems in Reactor Safety

1. Interaction of Fuel and Cladding with Liquid Sodium

a. Drop Experiments. Experiments are being conducted to determine the nature and magnitude of safety problems that might arise in the event that abnormally hot fuel and cladding materials come in contact with the coolant (usually sodium) in a fast reactor. Small heated samples of fuel or cladding materials were dropped into heated sodium in a survey study designed to discover unusual interactions, such as exothermic chemical reactions, extensive surface attack, frothing, disintegration, or alloy formation.

The present experiments consisted of heating and levitating (see Progress Report for May 1964, ANL-6904, p. 102) samples of the fuel or cladding (about 0.3 cc in volume) to temperatures as high as 2400°C and then dropping them into a container of liquid sodium. The quenched samples of reactor material were then removed from the sodium, cleaned with alcohol, and examined for evidence of interaction. The apparatus was housed in a glovebox containing an argon atmosphere in which the oxygen and water vapor concentrations were each maintained below 15 ppm.

In earlier work (see Progress Report for May 1965, ANL-7046, p. 81), molten samples of zirconium, vanadium, nickel, mild steel, and Type 304 stainless steel, and solid samples of niobium, niobium-1 w/o zirconium, molybdenum, and tantalum, were dropped into sodium at 250°C. No evidence of chemical interaction between these metals and sodium was found. The materials that were solid when dropped were simply quenched by the sodium. Fastax high-speed motion pictures taken during the tests showed that only moderate turbulence of the sodium surface occurred when the solid metal samples entered the sodium and that this was followed by a brief period of bubbling.

Several samples which were initially at temperatures greater than their melting points fragmented and caused violent eruptions of the sodium. Type 304 stainless steel caused the most vigorous effect as judged by the observed violence of the sodium eruption and fragmentation of the metal sample. Subsequently, several experiments were performed to investigate further the fragmentation of samples in sodium and to compare it with the effects obtained by quenching molten metals in water and other liquids such as molten potassium and lithium.

All experiments conducted to date on materials dropped into molten sodium are listed in Table XIX. It should be noted that the sample temperatures decreased about 100°C during the 2-ft drop into the sodium.

TABLE XIX. Results of Dropping Materials into 250°C Sodium
(Sample volume, ~0.3 cc; height of drop, 2 ft)

Material	Melting Point (°C)	Drop Temperatures at which the Samples Remained Intact (°C)	Drop Temperatures at which the Samples Fragmented (°C)
Tantalum	2980	1950, 2200, 2400	-
UO ₂	~2700	900, 2100	-
Molybdenum	2600	2200	-
Niobium	2470	2400	-
Niobium-1 w/o Zirconium	~2450	2200	-
Zirconium	1852	1700, 1860, 1900, 2000, 2100	2350
Titanium	1812	-	2050
Vanadium	1730	1800, 1950	2200
Vanadium-20 w/o Titanium	~1700	1800	2100
Nickel	1453	1400, 1600, 1800	2100
Mild Steel	~1450	1500, 1600	2000, 2400
Type 304 Stainless Steel ^a	~1400	1425	1550, 1600, 1800, 2000, 2100, 2200, 2300, 2400
Uranium	1133	1200	1700, 2000, 2450
Silver	961	-	1900
Aluminum	660	900	1200, 1550, 1950

^aStainless steel 304 samples were also dropped at a temperature of 2200°C into sodium maintained at 450, 600, 700, and 820°C; fragmentation occurred in all cases.

The effect of drop temperature on the degree of fragmentation is shown in Fig. 36. At 2200°C the stainless steel formed particles having an average diameter of about 10 mils.

Fig. 36. Residues of Type 304 Stainless Steel Samples at Various Initial Temperatures Dropped into Liquid Sodium

A number of experiments with stainless steel were performed at drop temperatures between 2200 and 2400°C, with reproducible results. The sequence of events resulting from a drop at 2400°C is shown in Fig. 37, where selected Fastax high-speed motion-picture frames illustrate the entrance of the drop, the resulting crater in the sodium, and the delayed violent ejection of small sodium particles (2-3 mm in diameter). In this experiment, about 15% of the stainless steel sample was also ejected during the violent period created by fragmentation of the sample.

Stainless steel samples were dropped not only the standard 2 ft from the levitation coil into the sodium, but were also dropped from 2-in. and 6-in. heights. The violence and extent of fragmentation did not appear to be affected by the differences in these drop distances. Therefore, impact velocity apparently was not a critical parameter.

Another series of experiments with stainless steel samples heated to 2200°C and dropped into sodium at 250, 450, 600, 700, and 820°C was performed to determine the effect of sodium temperature on the extent of fragmentation. The residues obtained in the experiments with sodium at 250, 450, and 600°C were similar in appearance and in the degree of fragmentation. The residue from the 700°C sodium experiment consisted primarily of shiny particles such as those obtained at lower sodium temperatures,

Approaching Na

$t = 0 \text{ msec}$

$t = 0.14 \text{ msec}$

$t = 0.48 \text{ msec}$

$t = 0.62 \text{ msec}$

$t = 0.76 \text{ msec}$

$t = 1.22 \text{ msec}$

$t = 3.58 \text{ msec}$

Fig. 37. Selected Fastax Frames of Type 304 Stainless Steel Sample Dropped into Molten Sodium
($T_{SS} = 2400^{\circ}\text{C}$; $T_{Na} = 250^{\circ}\text{C}$)

but contained some gray powder. The residue from the drop into 820°C sodium consisted entirely of gray powder; microscopic examination of this residue indicated that it consisted of very small metallic particles which adhered together in lumps. Analysis of the residue gave no evidence of chemical interaction with sodium, and X-ray diffraction studies showed only gamma (fcc) iron. The only effect of the higher sodium temperature was a more thorough dispersion.

Experiments were also conducted in which stainless steel samples at various temperatures were dropped into liquid potassium (250°C) and liquid lithium (275°C). Fragmentation occurred with both liquid metals at drop temperatures above the melting point of the stainless steel. The fragmentation in lithium was less extensive than in sodium, while the residues recovered from potassium were slightly more extensively broken up. Thus for these alkali metals the extent of fragmentation of the stainless steel increased with decreasing boiling point (b.p. Li, 186°C; Na, 98°C; K, 64°C).

Both uranium metal and UO₂ pellets were dropped into 250°C sodium. Uranium metal spheres fragmented over a wide range of drop temperatures from 1700 to 2450°C. The fragmentation was less violent than for the stainless steel. Although the stainless steel drops broke up into more or less spherical particles, the uranium samples formed a fragile filamentary network. Uranium dioxide when dropped as solid pellets did not fragment; however, extensive cracking occurred with a pellet initially heated to 2100°C. Sodium was absorbed into the cracks and was not removed by a brief alcohol rinse.

The principal finding of these experiments has been that violent fragmentation occurs with molten metals heated well above their melting points. This spontaneous fragmentation was surprising because it appeared to be contradictory to previous experience of dropping very hot molten metals into water. Molten pellets of zirconium, uranium, stainless steel, and aluminum dropped into water did not fragment.

An oxide film formed when these metals were dropped into water; this was thought to help prevent fragmentation of the samples in water. To test this hypothesis, spheres of noble metals were dropped both into water and into sodium. Although silver and gold do not form oxide films in water, these metals did not fragment when dropped into water; however, silver at 1900°C, when dropped into sodium, did suffer fragmentation. These samples are shown in Fig. 38. Thus it was concluded that the difference in behavior of the metal samples in water and liquid sodium is not caused by oxide films, but is due to differences in physical properties of water and sodium.

Fig. 38. Quenched Residues of Gold and Silver

A likely explanation for the fragmentation process is that the driving force for fragmentation might be the violent growth and collapse of sodium vapor bubbles which are characteristic of subcooled boiling in the transition and nucleate regimes. In general, it would be expected that the transition and nucleate boiling regimes would extend from nearly the boiling point to around the critical point. For sodium, this is a range of temperatures from 900 to 2300°C. All of the fragmentations occurred with molten metals cooling through some part of this temperature range. According to this hypothesis, fragmentation would also be expected to occur with molten metals in water, but only in the temperature region of violent boiling.

If this criterion for metal fragmentation is valid for water, then metals which have melting points below the critical temperature of water (373°C) should suffer fragmentation when the molten metals are quenched in water. Experiments were performed in which tin (m.p. 232°C), bismuth (m.p. 271°C), lead (m.p. 327°C), zinc (m.p. 420°C), and aluminum (m.p. 660°C), were heated to 825°C and dropped into water at 0°C. The results of several such experiments are shown in Table XX. Lead, bismuth, and tin suffered considerable fragmentation, whereas aluminum and zinc, as well as gold and silver, which have melting points above 373°C, did not fragment.

Experiments with Wood's Metal (m.p. 70°C) and Rose's Metal (m.p. 96°C) also fit the criterion properly; the samples when heated to 600°C and dropped into 0°C water fragmented. Further experiments with water indicated that subcooled boiling was necessary for fragmentation; thus, if the water temperature is raised to 60°C, tin, bismuth, and lead do not fragment. These same materials heated to 600°C and dropped into liquid nitrogen did not suffer fragmentation; this is likewise consistent with the fragmentation criterion, since nitrogen has a critical temperature of -147°C, which is well below the melting points of the metals.

TABLE XX. Results on Dropping Various Materials into Water
(Sample volume, ~0.3 cc; height of drop, 2 ft)

Material	Melting Point (°C)	Sample Behavior for Indicated Drop Temperature (°C)			
		Water at 0°C		Water at 60°C	
		Intact	Fragmented	Intact	Fragmented
Gold	1063	2600	-	- ^a	- ^a
Silver	961	1900	-	- ^a	- ^a
Aluminum	660	825	-	825	-
Zinc	420	825	-	825	-
Lead	327	-	825	825	-
Bismuth	271	-	825	825	-
Tin	232	-	825	825	-
Rose's Metal	96	-	600	- ^a	- ^a
Wood's Metal	70	-	600	- ^a	- ^a

^aNo experiments were done in 60°C water.

These preliminary studies of the interaction of reactor materials at temperatures well above their melting points with liquid sodium have identified a fragmentation mechanism that results in extensive subdivision of molten metals of interest to fast reactor technology. This knowledge should be useful in the interpretation of the results of in-pile meltdown tests of fast reactor fuel elements. It appears that chemical attack and alloy formation do not occur to a significant degree during the brief time available in a rapid quench with the reactor materials studied.

b. Penetration Experiments. In the Fast Reactor Safety Program, emphasis has been placed on safety-oriented studies designed to help explain fuel-rod failure during fast-reactor transients. Advanced metal fuels such as U-Pu-Zr and U-Pu-Ti have melting points below the cladding compositions of interest, such as Type 304 stainless steel and V-20 Ti alloy. Therefore these combinations are amenable to molten-penetration studies¹⁵ which establish rates of penetrations of claddings by molten fuel.

In this test, cladding specimens are dipped into a crucible of molten fuel at a known temperature and the time for the fuel to penetrate the specimen wall is recorded. One melt each of about 800-g size has been made of U-15 w/o Pu-10 w/o Zr and U-15 w/o Pu-6.5 w/o Ti alloy for the penetration medium. Specimens of cladding have also been machined and inspected by radiographic and ultrasonic methods to insure freedom from defects and nonuniform wall thickness.

¹⁵Walter, C. M., and Kelman, L. R., Penetration Rate Studies of Stainless Steel by Molten Uranium and Uranium-Fissium Alloy, J. Nucl. Matl. 6(3) 281-290 (1962).

V. PUBLICATIONS

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ANL-7088 SAMPLING OF LIQUID METALS

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L. Burris, Jr.ANL-7117 INSPECTION, EVALUATION, AND OPERATION OF THE
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N. Balai, C. R. Sutton, E. A. Wimunc, and R. F. Jones